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Natural Radionuclide Activity Concentrations and Radiological Hazard in Tea, Coffee, Wheat Fluor, and Powder Milk Consumed in Iraqi Markets

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Abstract. Twelve samples were collected, three samples for each type of tea, coffee, wheat flour and powder milk collected from Iraqi markets were analyzed by Sodium Iodide activated with Thallium NaI(Tl) detector to measure the concentrations of 137 Cs, 238 U, 232 Th, and 40 K in all samples. The average specific activity of 40 K were found to be 143.028±45.367 Bq/kg for coffee samples, 211.370 ± 15.413 Bq/kg for tea samples, 21.020 ± 2.540 Bq/kg for wheat samples, and 93.001 ± 24.580 Bq/kg for milk samples, respectively. The radiological hazards: radium equivalent, annual effective dose, internal and external hazard, life-time risk cancer, ingestion effective dose, and threshold consumption rate have been estimated, were all beneath the limit. The doses values were less than the limit value 0.30 mSv/y. Therefore, the consumption of these foods has no health risks. This process may help to obtaining basics on radiological health regulations.

INTRODUCTION

Updating the knowledge and analyzing the effects of exposure to radiation and its hazard are very important now a day. Coffee, tea, wheat flour, and milk powder are generally used in Iraq in particular, and its consumption is increasing. ⁴⁰K, ²³²Th and ²³⁸U causes a danger to the human body if it exists inside it where the presence of irradiation of organs and tissues of the cells of the body [1].

However, annual consuming of foods varies across countries. In addition, coffee, tea, and wheat are consuming in several places of the world [2].

Milk is an important food for human nutrition; it is beneficial in the human diet and mostly needed by infant, children, and adults during their growing age. ⁴⁰K decays by the beta emitter (89% of the times) to ⁴⁰Ca and decays by gamma emitter (11% of the times) to the gas ⁴⁰Ar. So, ⁴⁰K can present both external health hazards from gamma radiation energy ($E\gamma = 1.46$ MeV) and internal health hazards from beta particles (EMax = 1.35 MeV); while inside the body, ⁴⁰K poses a health risk from the emission of, gamma rays associated by cell damage and a general potential for cancer induction subsequent [3].

Some natural radionuclide and other elements found in the ground are absorbed by plants through the roots to be transmitted to the plant itself [4]. This transition is carried out in a distributed manner on all the tissues of the plant, depending on properties of soil and the plant [5]. Most natural materials contain an acceptable amount of natural radioactivity of ²³⁸U, ²²⁶Ra, ²³²Th, ⁴⁰K, etc and the most common is potassium [6].

Many literatures conducted the evaluation of activity of the natural isotopes in different kinds of selected materials to guess the toxicity of the effectiveness of these concentrations [7, 8, 9, 10, 11].

This paper aimed to estimate the activities of ²³⁸U, ²³²Th, and ⁴⁰K in coffee, tea, wheat flour, and milk powder samples, measured by Na(Tl) detector, To enrich the basic information about natural radioactive materials its focus and presence in Iraq in particular in food is infrequent. For this study, the twelve samples were measured

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of which three samples for each kind have been selected. Activities including ingestion effective dose have been calculated, so as to compare the results with the recommended limits. The materials are thus evaluated.

MATERIAL AND METHOD

Collection and Preparation of Samples

Twelve samples were collected. Three samples for each type of tea, coffee, wheat flour and powder milk available in the local Iraqi market and consumed by citizens to measure the natural radiation activity. The samples were dried and crushed with mill to obtain a homogeneous powder. After the grinding process, the powder was sifted through a sieve of 630 microns. After that, the samples were filled in Marinelli Becker and sealed with provisions. The code name for each sample was installed on Marinelli Beckers. Marinelli Beckers were closed for 30 days prior to the screening process in order to achieve a secular equilibrium. **Table 1** listed the name and origin of the studied samples.

Radioactivity Measurement

Detection of ²³⁸U, ²³²Th, and ⁴⁰K in coffee, tea, wheat flour, and milk powdered samples were carried out with gamma spectrometry technique using 3×3 inch NaI(Tl) detector, Alpha Spectra Inc., scintillation detectors, Model 12/12/3, Serial (031215G), Made in United States of America (U.S.A). The detector had 90% efficiency and energy resolution of 28.74 keV at 1332 keV of ⁶⁰Co.

The multichannel analyzer used is a digital analyzer of type Bright SPEC model bMCA (blug-on Multichannel analyzer. It analyzes the gamma spectrum for gamma rays at 4096 channels. Note that this analyzer can be controlled and changing the characteristics and number of channels through a specialized computer program (bMCA). It contains a red light indicator for the intermittent gamma-rays to the detector and it coupled to a computer by USB cable to transmit the signal to bMCA program and finally, display the analyzed spectrum on the screen.

Standard assured quality samples were used for calibration and absolute efficiency (obtained from IAEA) for calibration and absolute efficiency. Combination of radionuclides, with corresponding energies, include ¹³³Ba (81 and 356.01 keV), ⁶⁰Co (1173.24 and 1332.50 keV), and ¹³⁷Cs (661.66 keV). The measurement time for radiological back-ground and samples took 86,400 seconds. ²³⁸U activity was obtained by gamma line by the decomposition of the product ²¹⁴Bi (1764.5 keV). ²³²Th activity of the weighted average activities of gamma peaks of ²⁰⁸Tl (583.19 and 2614.5 keV) was considered. Concentrations of ⁴⁰K and ¹³⁷Cs were evaluated through identify γ -lines characteristic of energies, 1460.8 and 661.61 keV respectively. In addition, the minimum average detectable activities of ²³⁸U, ²³²Th, ⁴⁰K, and ¹³⁷Cs using NaI(Tl) detector were 1.628, 0.155, 18.797, and 0.128 Bq/kg, respectively.

No.	sample	Name of sample	Exporting country
1		Al-Dhiafa	Iraq
2	Coffee	Hamwi	Emirates
3		Winner	Syria
4		Ahmed	UK
5	Tea	Al-Wazah	South of Sri Lanka
6		Mahmood	India
7		Durra	Jordan
8	Wheat	Jenan	Emirates
9		Karbala	Turkey
10		Anchor	New Zealand
11	Milk	Dielac	New Zealand
12		Al-Mudhish	Sultanate Oman

Table 1. Name of sample and exporting country of samples.

CALCULATIONS

Specific Activity

The activity specified is the activity per unit block, the unit used is either Ci/kg or Bq/g. It can be calculated as [12]:

$$A(E_{\gamma}) = \frac{N}{\epsilon(E_{\gamma}) \times I_{\gamma}(E_{\gamma}) \times t \times m}$$
(1)

Where: N is the counting of gamma rays (i. e. area under the photo peaks) and m is the sample mass in kg.

Absorbed Dose Rate D

According to the United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR, 1988 and 2010 [13, 14], it can be given by:

$$D_{out}(nGyh^{-1}) = 0.427 A_U + 0.662A_{Th} + 0.043 A_K$$
(2)

Where A_U , A_{Th} and A_K are the concentrations of ²³⁸U, ²³²Th, and ⁴⁰K in ($Bqkg^{-1}$) respectively. While D_{in} can be calculated as follows, provided by UC European Commission, 1999 [15]:

$$D_{in}(nGy h^{-1}) = 0.92 A_U + 1.1A_{Th} + 0.081 A_K$$
(3)

Radium Equivalent Activity Raeq

For uniformly distributed the Raeq estimated from UNSCEAR, 2000 [16]:

$$Ra_{eq}(Bq, kg^{-1}) = A_U + 1.43 A_{Th} + 0.077 A_K$$
(4)

The rate of γ -dose limit of Ra_{eq} is 370 $Bqkg^{-1}$ recommended by UNSCEAR, 2000 [16].

Hazard Index H

 H_{ex} is obtained from the following, UNSCEAR, 2000 [16]:

$$H_{e_{\mathcal{K}}} = \frac{A_U}{370 \ Bq.kg^{-1}} + \frac{A_{Th}}{259 \ Bq.kg^{-1}} + \frac{A_{\mathcal{K}}}{4810 \ Bq.kg^{-1}} \tag{5}$$

While H_{in} is as [16]:

$$H_{in} = \frac{A_U}{185 \ Bq.kg^{-1}} + \frac{A_{Th}}{259 \ Bq.kg^{-1}} + \frac{A_K}{4810 \ Bq.kg^{-1}}$$
(6)

Provided that for safety consume the upper limit should be < 1, as reported by UNSCEAR, 2000 and ICRP, 2007 [16, 17].

Annual Effective Dose Equivalent (AEDE)

AEDE is given as [16]:

$$AEDE_{out}(\mu Sv/y) = D_{out}(nGy/h) \times 8760(h/y) \times 0.20 \times 0.7(Sv/Gy) \times 10^{-3}$$
(7)

$$AEDE_{in}(\mu Sv/y) = D_{in}(nGy/h) \times 8760(h/y) \times 0.80 \times 0.7(Sv/Gy) \times 10^{-3}$$
(8)

Life-Time Cancer Risk (ELCR)

The **ELCR** risk is given as [18]:

$$ELCR_{out} = AEDE_{out} \times DL \times RF \tag{9}$$

$$ELCR_{in} = AEDE_{in} \times DL \times RF \tag{10}$$

Where *DL* is the duration of life, 70 years, and *RF* is a fatal cancer risk factor, 0.05/Sv, as defined by ICRP, 2012 [19].

Dose Ingested

The Dose Ingested is given by [20, 21]:

$$AACED (Sv/y) = CR \times SDCF_i \times A_i$$
(11)

Where, *CR* is the radionuclide consumption rate, *SDCF*_i are factors for adults given in refs [16, 22], and A_i is as defined in equation (1) for each radionuclide *i*. The annual threshold of *CR* (kg/y) is given by:

$$CR = \frac{3 \text{ AACED}}{\sum_{i=1}^{3} (SDCF_i \times A_i)}$$
(12)

RESULTS AND DISCUSSION

²³⁸U, ²³²Th, and ⁴⁰K specific activities in coffee, tea, wheat flour and powdered milk imported to Iraqi were evaluated. Table 2 tabulated the results for coffee; the activity of ²³⁸U ranged from 1.628 to 8.341 Bq/kg with average value 4.887 \pm 3.360 Bq/kg and from 0.528 to 3.206 Bq/kg for ²³²Th, respectively. In addition, the specific activity of ⁴⁰K varied from 93.812 to 183.177 Bq/kg with average of 143.028 \pm 45.367 Bq/kg and 0.269 to 0.483 Bq/kg with mean value 0.395 \pm 0.112 Bq/kg, for ¹³⁷Cs. For tea, the average activity concentrations are 6.541 \pm 2.140, 0.951 \pm 0.175, 2.742 \pm 0.749, and 211.370 \pm 15.413 Bq/kg , for wheat 4.803 \pm 0.940, 0.950 \pm 0.292, 1.478 \pm 0.493, and 21.020 \pm 2.540 Bq/kg , for milk are 3.392 \pm 2.522, 0.499 \pm 0.582, 2.025 \pm 1.004, and 93.001 \pm 24.580 Bq/kg. for ²³⁸U, ²³²Th, and ⁴⁰K, respectively. While the average activity concentration of ¹³⁷Cs for all samples was lower than the unit. All results are less than the global limits (33, 45, and 412 Bq/kg for ²³⁸U, ²³²Th, and ⁴⁰K respectively) reported by UNSCEAR, 2010 [14].

Figures 1, 2, 3, and 4 show the activity concentrations of 40 K in Al-Wazah tea sample which had the highest activity concentration 224.687 Bq/kg, while Jenan sample of wheat had the lowest value 18.797 Bq/kg. For all samples, the activity concentration values of 40 K were less than 412 Bq/kg as recommended by UNSCEAR, 2010 [14].

Moreover, the current results from wheat flour showed average concentration of 238 U was 3.18 times higher than the results obtained by Ali et al., 2014 [10]; and the average concentration of 232 Th and 40 K was lower than the average concentration found in the present study.

In addition, the results of milk powder the concentration of 238 U obtained by Ali et al., 2015 [23] was 4.5 times higher than the present study, no difference for 232 Th concentration, and the concentration of 40 K obtained by Ali et al., 2015 [23] was 3.20 times higher than the present study.

Also, when comparing the results of the activity concentrations in the milk samples the concentrations of uranium and thorium were found to be in good compatibility with Hassan et al., 2017 results [24]. While the ⁴⁰K concentration found by Hassan was 1.91 times higher than the current study.

Tables 2 also listed the calculated values of radium equivalent activity. The results showed that the average values of radium equivalent were 16.587 ± 4.201 , 24.177 ± 3.411 , 7.780 ± 1.105 , and 11.267 ± 5.195 Bq/kg for coffee, tea, wheat, and milk respectively. These results are < 370 Bq/kg [16].

Table 3 shows the results of radiation indices for all samples. All the radiation hazard indices values were below the permissible limits of recommended by UNSCEAR, 2000 [16] and as prescript by UNSCEAR, 2010 [14]. Table 3 and Figure 5 show a variation in the estimation of Life-time cancer risk. Its value within the body was higher than that outside the body. The highest risks in and out of the body, was found in the Al-Wazah tea sample. The same criteria was followed by the Hamwi coffee sample. The highest cancer risk in milk samples was in Anchor milk; while in Wheat the highest cancer risk was in Karbala sample.

Table 4 and Figure 6 show the estimated activities of radionuclides ²³⁸U, ²³²Th, and ⁴⁰K from each sample. All values of AACED were below the recommended limit 0.3 mSv/yr, for ingestion dose, as reported by UNSCEAR, 2010 [14]. The dose ingested is changing from 0.013 mSv/y in Jenan wheat to 0.122 mSv/y in Al-Wazah tea, respectively. The highest mean value 0.0114±0.009 mSv/y was recorded in tea samples, while the minimum mean value 0.014±0.001 mSv/y was in wheat samples. These results pointing that the doses ingested were < 1 mSv/y [20].

Sample	Name	Specific activity concentrations (Bq/kg)					
		²³⁸ U	²³² Th	²³² Th	⁴⁰ K	¹³⁷ Cs-137	Ra eq
		(1764.5	(2614.5	(583.19	(1460.8 keV)	(661.61	
		keV)	keV)	keV)	,	keV)	(Bq/kg)
Coffee	Al-Dhiafa	1.628	0.528	3.206	183.177	0.483	16.488
	Hamwi	8.341	0.548	2.620	152.096	0.433	20.835
	Winner	4.691	0.364	1.149	93.812	0.269	12.436
	Average	$4.887 \pm$	$0.480 \pm$	$2.325 \pm$	$143.028 \pm$	$0.395 \pm$	
	-	3.360	0.101	1.060	45.367	0.112	$16.587 {\pm} 4.201$
Tea	Ahmed	5.492	0.971	2.147	214.938	0.421	23.431
	Al-Wazah	9.004	1.115	2.495	224.687	0.244	27.810
	Mahmood	5.128	0.768	3.583	194.486	0.475	21.201

Table 2. Specific Activity concentrations for (²³⁸U, ²³²Th, ⁴⁰ K and ¹³⁷Cs) and Radium equivalent activities of samples.

	Table 2. continued Specific activity concentrations (Bq/kg)						
Sample	Name	²³⁸ U	²³² Th	²³² Th		¹³⁷ Cs-137	Ra eq
		(1764.5	(2614.5	(583.19	⁴⁰ K	(661.61	•
		keV)	keV)	keV)	(1460.8 keV)	keV)	(Bq/kg
	Average	$6.541\pm$	$0.951 \pm$	$2.742 \pm$	$211.370 \pm$	$0.380 \pm$	
		2.140	0.175	0.749	15.413	0.121	24.177 ± 3.411
	Durra	3.943	0.623	1.232	23.789	0.918	6.666
Wheat	Jenan	4.658	1.184	1.155	18.797	0.658	7.799
	Karbala	5.807	1.043	2.046	20.476	0.822	8.875
	Average	$4.803~\pm$	$0.950 \pm$	$1.478 \pm$	$21.020 ~\pm$	$0.799 \pm$	
	-	0.940	0.292	0.493	2.540	0.131	7.780 ± 1.105
	Anchor	6.304	1.171	2.058	120.249	0.261	17.237
Milk	Dielac	1.896	0.170	1.004	86.255	0.128	8.782
	Al-Mudhish	1.977	0.155	3.012	72.499	1.012	7.781
	Average	$3.392 \pm$	$0.499 \pm$	$2.025 \pm$	$93.001 \pm$	$0.467 \pm$	$11.267 \pm$
		2.522	0.582	1.004	24.580	0.476	5.195
Permissible Limit		33	4	-5	412		370
UNSCEAR, 2010							UNSCEAR,
							2000

	D (nGy/h)		AEDE (µSv/y)		Hazard index		ELCR	
Name	outside	inside	Outside	inside	Hex	Hin	outside	Inside
Al-Dhiafa	8.922	16.917	10.942	82.986	0.045	0.049	38.295	134.034
Hamwi	10.464	20.596	12.833	101.035	0.056	0.079	44.917	157.209
Winner	6.278	12.315	7.700	60.414	0.034	0.046	26.949	94.319
Ahmed	12.230	23.531	14.999	115.432	0.063	0.078	52.497	183.739
Al-Wazah	14.245	27.710	17.470	135.935	0.075	0.099	61.144	214.003
Mahmood	11.061	21.316	13.565	104.566	0.057	0.071	47.477	166.171
Durra	3.119	6.240	3.826	30.612	0.018	0.029	13.389	46.863
Jenan	3.581	7.110	4.392	34.881	0.021	0.034	15.372	53.801
Karbala	4.051	8.149	4.968	39.973	0.024	0.040	17.387	60.855
Anchor	8.637	16.827	10.593	82.548	0.047	0.064	37.075	129.763
Dielac	4.631	8.919	5.680	43.751	0.024	0.029	19.880	69.580
Al-Mudhish	4.064	7.862	4.984	38.566	0.021	0.026	17.446	61.059

Table 4. AACED ingestion dose of radionuclides ²²⁶Ra, ²³²Th, and ⁴⁰K and the rate of consumptions, for studied samples.

Samples	Name	AACED	Threshold
		for 1 kg/year	consumption rate
		(mSv/year)	(kg/year)
	Al-Dhiafa	0.096	36.7276
Coffee	Hamwi	0.084	22.4609
	Winner	0.051	40.1213
	Average	0.077 ± 0.024	33.1034 ± 9.3714
	Ahmed	0.115	25.0875
Tea	Al-Wazah	0.122	18.9692
	Mahmood	0.105	24.7085
	Average	0.114 ± 0.009	$22.9217{\pm}3.4282$
	Durra	0.015	53.6272
Wheat	Jenan	0.013	45.9456
	Karbala	0.015	36.5364
	Average	0.014 ± 0.001	$45.3697 {\pm}\ 8.5599$
	Anchor	0.066	27.6654
Milk	Dielac	0.046	67.3691
	Al-Mudhish	0.040	51.9792
	Average	0.051 ± 0.014	49.0046 ± 20.0183



FIGURE 1. Specific activity of coffee samples.



FIGURE 2. Specific activity of tea samples.



FIGURE 3. Specific activity of wheat flour samples.



FIGURE 4. Specific activity of powder milk samples.



FIGURE 5. Life-time cancer risk of samples.



FIGURE 6. Ingestion effective dose of samples.

CONCLUSION

The ²³⁸U, ²³²Th, and ⁴⁰K activity concentrations in tea were greater than those in coffee, wheat and milk and all of them were lower than the allowable value. The estimation of the average risk indicators are safe for radiation hazards.

Hence the studied samples are safe to be consumed. Therefore, all the results of the samples under study indicate that the radiation hazard associated with drinking coffee are moderate compared to drinking tea. Adults may face a greater health risk when they drink more tea instead of coffee because they are more likely to accumulate. While the ingestion effective dose for wheat and milk samples are far below the limit.

Therefore, the analysis confirms that the studied samples have no significant radioactive effects. Thus the quality control must be carried out since the study of the concentration of radionuclides in this field is of great importance. Therefore, in terms of market, it is preferable to determine the values of radioactivity of the commodity important to the population in general.

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