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Radium and Uranium Concentrations Measurements in Vegetables Samples of Iraq

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Abstract

In the present study twenty-two vegetable samples were collected from Iraqi market. Sealed can technique using CR-39 plastic track detector strippable has been used in order to measure radium and uranium concentrations. Etching was done with 6.25 N NaOH and optical microscope was used with the purpose of counting of alpha particle tracks. The values of effective radium content are found to range from 0.074 Bq/ kg to 0.566 Bq/ kg with the mean value of 0.317 Bq/kg. The values of uranium concentrations are found to range from 0.081 ppm to 0.615 ppm with the mean value of 0.345 ppm. Positive correlation has been observed between radium concentration and uranium concentrations in vegetable samples. Measurements of radium and uranium concentrations in vegetables are important from the health protection point of view, so simple and reliable analytical methods must be available.

Keywords

Radium, Uranium, Can Technique, CR-39 Detectors, Vegetables, Iraqi Markets

1. Introduction

Studies on radiation levels and radionuclide distribution in the environment provide vital radiological baseline information. Such information is essential in understanding human exposure from natural and man-made sources of radiation and necessary in establishing rules and regulations relating to radiation protection [1].

Uranium and radium belong to the group of primordial radionuclides, as they have always been present in the earth. The radionuclides ^{238}U, ^{235}U and ^{232}Th, which decay through three distinct series of radionuclides, are of great importance in the nuclear fuel cycle. Not only are they present in the human body and foodstuffs but some

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gaseous radionuclides may also be inhaled. ^{226}Ra (half-life 1600 years), ^{224}Ra and ^{228}Ra (half-lives 3.6 days and 5.8 years, respectively, and both usually mixed with uranium ore) are of radiological importance because radium behaves chemically like calcium, being deposited on bone surfaces and areas of mineral metabolism [2].

^{226}Ra in the environment is widely distributed, being present in various concentrations in waters, soils, sediments and rocks [3]. When radium is ingested, the majority of material is rapidly excreted. However, since the chemical behavior of radium is similar to that of calcium, radium absorbed to blood from the GI-tract or lungs follows the behavior of calcium and is primarily deposited in bone [4]. Radium is a common radionuclide in the environment and it is the parent of radon. Its form is the most deadly radionuclide's because it produces alpha radiation and has a very long half-life [5]. The immediate radon precursor is radium (^{226}Ra) that spreads widely, particularly in materials which are made from mineral products. The forerunner of radium is Uranium (^{238}U); which has a half-life of 4.47×10^9 years [6]. During the last decade, there has been an increasing interest in the study of radium activity in various vegetables and food. Since radium is a highly radioactive chemical element and it is the most important source of radioactivity in a variety of foodstuffs. Radium is a solid radioactive element under ordinary conditions of temperature and pressure [7]. Microscopic quantities of radium in the environment can lead to some accumulation of radium in bone tissue whereby it degrades bone marrow and can mutate bone cells. Ingestion or body exposure to radium causes serious health effects which included sores, anemia, bone cancer and other disorders [8].

Radium present in the soil is taken by the plants and enters into the body with vegetarian/non-vegetarian food and tends to follow calcium metabolic process to become concentrated in bones [9].

Uranium has been detected in a variety of foodstuffs. The highest concentrations are found in shellfish, and lower levels have been measured in fresh vegetables, cereals and fish. The average per capita intake of uranium in food has been reported to be 1.3 μg/day [10] and 2 - 3 μg/day [11] in the USA and 1.5 μg/day in Japan [12].

In a review of naturally occurring sources of radioactive contamination in food, dietary intakes of ^{238}U were found to range from 12 to 45 mBq/day in several European countries, from 11 to 60 mBq/day in Japan (the higher values were found in uranium mining areas) and from 15 to 17 mBq/day in the USA. The average daily dietary intake was in the order of 20 mBq, or about 4 μg. It was often difficult to determine whether these dietary intakes included intake from drinking-water, and it was emphasized that intake from drinking-water has sometimes been found to be equal to intake from the diet [13].

Following ingestion, uranium rapidly appears in the bloodstream [14], where it is associated primarily with the red cells [15]; a non-diffusible uranyl-albumin complex also forms in equilibrium with a diffusible ionic uranyl hydrogen carbonate complex ($UO_2HCO_3^+$) in the plasma [16].

Because of their high affinity for phosphate, carboxyl and hydroxyl groups, uranyl compounds readily combine with proteins and nucleotides to form stable complexes [16]. Clearance from the bloodstream is also rapid, and the uranium subsequently accumulates in the kidneys and the skeleton, whereas little is found in the liver [14]. The skeleton is the major site of uranium accumulation [17]. Based on the results of studies in experimental animals, it appears that the amount of soluble uranium accumulated internally is proportional to the intake from ingestion or inhalation. It has been estimated that the total body burden of uranium in humans is 40 μg, with approximately 40% of this being present in the muscles, 20% in the skeleton and 10%, 4%, 1% and 0.3% in the blood, lungs, liver and kidneys, respectively [18].

Uranium is the proximate source of radium and radon in the soil and rocks. Uranium prospection through the analysis of soil, rocks, plants and water has been reported by many workers [19] [20]. Uranium, present in the earth, is transferred to water, plants, food supplements and then to human beings. Uranium accumulated in humans may have a dual effect due to its chemical and radioactive properties. High intake of uranium and its decay products may lead to harmful effects in human beings. According to an estimate [21], food contributes about 15% of ingested uranium, while drinking water contributes about 85%. An exposure of about 0.1 $mg \cdot kg^{-1}$ of body weight of soluble natural uranium results in transient chemical damage to the kidneys [22].

The aim of this research was to determine uranium and radium concentrations in twenty two types of vegetables. So, the measurement of uranium and radium concentrations in vegetables is necessary to investigate the role of radium and uranium concentration in causing various diseases, especially cancer.

2. Materials and Method

In the present study, 22 different samples from vegetables, were investigated. The samples were collected from

Iraqi market. Vegetable samples were washed, peeled when necessary, and dried in air. After that, they were oven dried at 80˚C for approximately 16 h [23].

The vegetable samples were dried in oven, minced, crushed, sieved by 1-mm mesh, weighted, and carefully sealed for 81 days in a cylindrical containers made of plastic with dimensions of 3.5 cm in diameter and 6.5 cm in depth. A piece of CR-39 detector of 500 µm thicknesses (American Technical Plastic, Inc.) with area of (1.5 × 1.5) cm was embedded in middle of samples (closed contact α-autoradiography) in each container. The detector records the tracks of alpha-particles emitted by radon gas produced through the alpha-decay of radium contents of the samples. After their radiation period, the bombarded detectors were collected and chemically etched in NaOH solution 6.25 M at 70˚C during 8 h [24]. After etching, the CR-39 detectors were washed in distilled water and then dipped for few minutes in a 3 % acetic acid solution and washed again with distilled water and dried in air. Can technique using CR-39 detectors have been used for measurements of uranium and radium concentration [25] as shown in **Figure 1**. The background of CR-39 track detectors were counted by optical microscope and subtracted from the count of all detectors. The tracks density in the nuclear track detector was counted using an optical microscopic (kruss-mbl2000) at a magnification of 400×.

3. Results and Discussion

The "can technique" proposed by Alter and Price [26] and then developed by Somogyi [27] was used to calculate the radium and uranium concentrations in vegetables samples. The radium concentration in vegetable has been calculated by using the relation:

$$C_{\mathrm{Ra}} = \frac{\rho h A}{K T_e M} \tag{1}$$

where C_{Ra} is the effective radium content of vegetables samples (Bq/kg), M is the mass of vegetables sample (0.0185 kg), A is the area of cross-section of the bottle (9.6211×10^{-4} m^2), h is the distance between the detector and the top of the vegetable sample (0.049 m), K is the sensitivity factor, which is equal to 0.045987 tracks·cm^{-2}·d^{-1} per Bq·m^{-3} and T_e is the effective exposure time, which is related to the actual exposure time T and decay constant λ for ^{222}Rn by the relation [28].

$$T_e = T - \frac{1}{\lambda}\left(1 - \mathrm{e}^{-\lambda T}\right) \tag{2}$$

uranium concentrations C_U in units of part per million (ppm) of vegetable samples has been calculated using the following equation [29]:

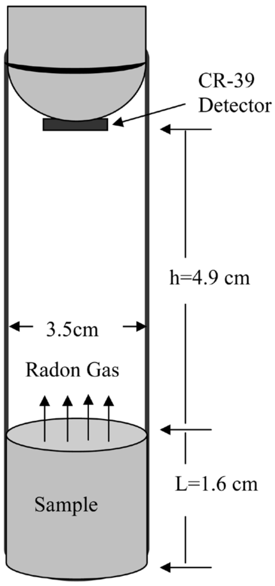

Figure 1. Shows a test tube technique used in the study.

$$C_U\left(\text{ppm}\right)=\frac{W_U}{W_S} \quad (3)$$

where W_S *is* the weight of sample (18.5 gm).

Uranium weight (W_U) in sample can calculate from the following equation [Richard and Josef, 1995] [30]:

$$W_U\left(\text{gm}\right)=\frac{N_U W_{mol}}{N_{Av}} \quad (4)$$

where N_U : numbers of uranium atoms, W_{mol} : weight molecular uranium, N_{Av} : number of Avogadro 6.023 × 10^{23} atom/mol.

The results of radium and uranium concentrations from the vegetables samples are presented in **Table 1**. **Figure 2** shows the relation between uranium concentration with the radium concentrations *i.e.* positive correlation has been observed between uranium concentration and radium concentration in vegetable samples of this study.

The Eggplants were characterized by the highest value of radium concentration 0.566 Bq/kg, while the lowest value of radium concentration in vegetables was found in the Parsley 0.074 Bq/kg. In addition to that, the average value of radium concentration in vegetables was 0.317 Bq/kg. The uranium concentrations are found to vary from 0.081 to0.615 ppm with a mean value of 0.345ppm for the same samples.

From **Table 1**, it has seen observed that there are variations in the values of uranium and radium concentrations among the samples. This variation may be arisen due to the difference in the nature of the samples and nuclei content of this samples.

Table 1. Radium and uranium concentrations in vegetable samples.

No. of samples	Vegetable samples	Code	ρ (Track/cm^2)	C_{Ra} (Bq/Kg)	W_U (μgm)	C_U (ppm)
1	Green Pepper	GP	200.892	0.147	2.962	0.160
2	Red Radish	RR	374.698	0.275	5.525	0.299
3	Cauliflower	CF	399.527	0.293	5.891	0.318
4	Onion	ON	349.869	0.256	5.159	0.279
5	White Radish	WH	275.381	0.202	4.061	0.220
6	Potato	PO	300.210	0.220	4.427	0.239
7	Chard	CD	225.722	0.165	3.328	0.180
8	Parsley	PY	101.579	0.074	1.497	0.081
9	Fenugreek	FK	151.234	0.111	2.230	0.121
10	Spinach	SH	622.992	0.457	9.187	0.497
11	Celery	CY	697.480	0.511	10.285	0.556
12	Basil	BL	449.186	0.329	6.624	0.358
13	Leek	LK	325.039	0.238	4.793	0.259
14	Cabbages	CS	126.404	0.092	1.864	0.101
15	Carrots	CA	424.357	0.311	6.275	0.338
16	Cress	CE	548.504	0.402	8.088	0.437
17	Eggplants	ES	771.968	0.566	11.384	0.615
18	Cucumber	CR	523.674	0.384	7.722	0.417
19	Mallow	MW	598.163	0.439	8.820	0.477
20	Lettuce	LE	647.821	0.475	9.553	0.516
21	Green Okra	GO	672.651	0.493	9.919	0.536
22	Turnips	TS	747.139	0.548	11.017	0.596
	Minimum		101.579	0.074	1.497	0.081
	Maximum		771.968	0.566	11.384	0.615
	Mean		433.385	0.317	6.391	0.345

The determination of radium and uranium concentrations in vegetable samples is very important in the exploration of the natural resources of these elements.

We found the uranium content in the vegetable samples was low and not significant from a health hazard point of view.

Figure 3 and Figure 4 have shown the distribution of radium and uranium concentration respectively, for different vegetable samples in Iraqi market.

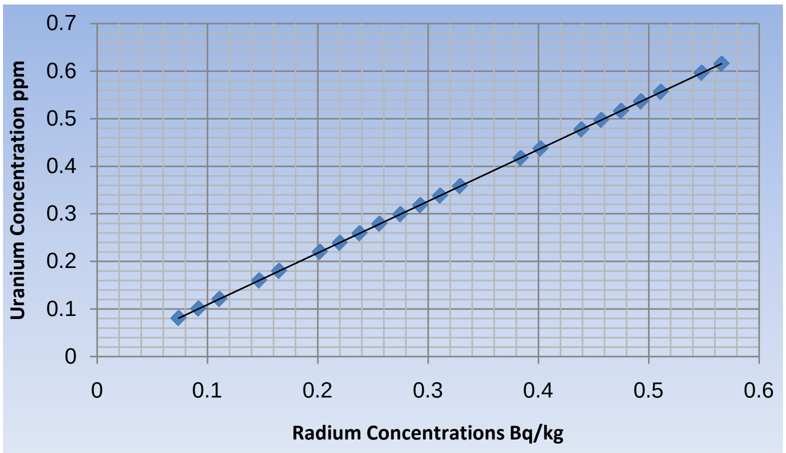

Figure 2. Showing the correlation between radium and uranium concentrations.

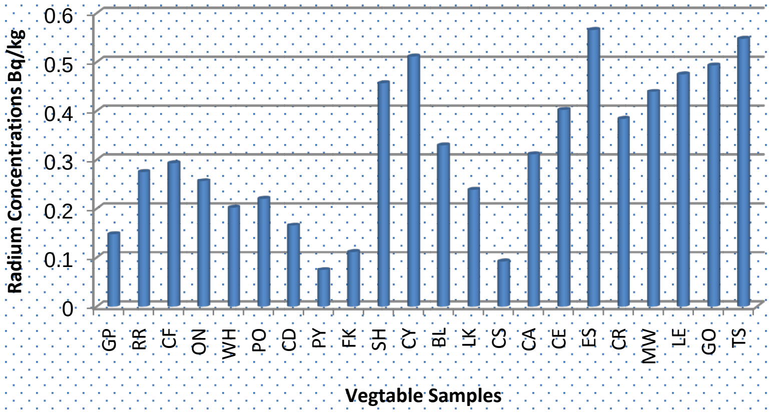

Figure 3. Radium concentrations in twenty two vegetable samples.

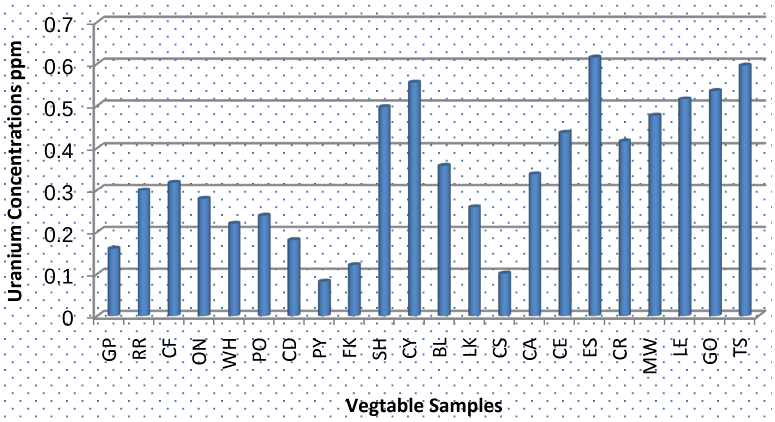

Figure 4. Uranium concentrations in twenty two vegetable samples.

The result of the uranium concentration of vegetable samples is quite low compared with the allowed limit (11.7 ppm) [31].

In Egypt, Ibrahim *et al*. (2007) reported radium content in twelve types of fresh vegetables were investigated for ^{226}Ra and the results are listed in **Table 2**. It was found that the maximum level of ^{226}Ra was 2.11 ± 0.01 Bq/kg detected in garden rocket, while the minimum level of <0.32 Bq/kg was detected in carrot, cucumber, green haricot, green bean, and spinach [32]. The values of effective radium content presented in this paper for vegetables samples are lower than those reported by Ibrahim *et al*. (2007) for garden rocket and agree with the values for carrot, cucumber, green haricot, green bean, and spinach [32].

In India, Shanthi *et al.* (2009) [33] shows that the activity concentration of ^{226}Ra for vegetables varies between 0.064 ± 0.03 (tomato) and 1.227 ± 0.24 Bq/kg (drumstick).

The maximum values of effective radium content in the vegetables samples studied are below the maximum than those reported by Shanthi *et al*. 2009, while the minimum value in vegetables samples are higher than values for the same study.

Concentrations of radium in foods vary widely because of the differing background levels, climate, and agricultural conditions that prevail. There are also differences in the types of local foods that may be included in the categories such as vegetables and fruits. The database is summarized in **Table 3** [31]. The values of radium content reported in **Table 3** are less than the values presented in this study for vegetables samples except maximum values in Germany.

Table 2. The levels of ^{226}Ra in fresh vegetables [32].

Type of samples	^{226}Ra Bq/kg
Carrot	0.32<
Cucumber	0.32<
Garden rocket	2.11 ± 0.01
Green haricot	0.32<
Green bean	0.32<
Mallow	0.63 ± 0.17
Lettuce	1.05 ± 0.48
Green okra	0.86 ± 0.05
Potato	0.80 ± 0.49
Spinach	0.32<
Squash	0.62 ± 0.25
Tomato	0.96 ± 0.30

Table 3. The levels of ^{226}Ra in leafy vegetables and root vegetables and fruits [31].

Leafy vegetables		Root vegetables and fruits	
Country	^{226}Ra (Bq/kg)	Country	^{226}Ra (Bq/kg)
United States	0.056	United States	0.007 - 0.047
China	0.075	China	0.063
Germany	0.006 - 1.150	Japan	0.011
Italy	0.027 - 0.044	Germany	0.005 - 9.400
Poland	0.037 - 0.043	Italy	0.014 - 0.025
U.K	0.022 - 0.170	Poland	0.011 - 0.215
-	-	U.K	0.009 - 0.041
-	-	Romania	0.009 - 0.190

4. Conclusions

The concentrations of radium and uranium have been measured using can technique method. The results showed that radium and uranium concentration in vegetable samples were ranged between (0.074 - 0.566) Bq/kg and (0.081 - 0.615) ppm, respectively. In addition, the highest concentration was found in Eggplants sample, whereas the lower concentration was found in the Parsley sample.

The results also show that, the mean values of radium and uranium concentrations for all vegetable samples are equal, 0.317 Bq/kg and 0.345 ppm, respectively.

The result of the uranium concentration of vegetable samples is quite low compared with the allowed limit (11.7 ppm). The radium distribution is found to be heterogeneous as the radium content in the vegetables samples varies from sample to another.

The results have revealed that the samples were safe as far the health hazard effects are concerned. Positive correlation has been observed between radium content and uranium concentrations in vegetable samples.

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