







# DETERMINING THE RADIUM CONCENTRATION IN VEGETABLES AND FRUITS IN AI-NAJAF, IRAQ

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↳Supporting Information

**ABSTRACT:** Amount of pollution radiation in foodstuffs and feedstuffs are very necessary to measure because it is a direct contact with human and animal health. Therefore, the present investigation is useful for the health and environmental data base. The study included measurement of effective radium-226 content ( $C_{Ra}$ ) in some chosen samples of vegetables and fruits (local and imported) in Najaf governorate. The  $C_{Ra}$  were measured in samples of vegetables and fruits using Solid State Nuclear Track Detectors (LR-115 Type II). Also, the annual effective dose (AED) associated with the exposure due to annual intake of  $^{226}Ra$  were calculated from ingestion of vegetables and fruits samples for adults. The results were revealed that the average value of  $C_{Ra}$  in vegetables and fruits samples in the present study was  $3.98 \pm 1.08$  Bq/kg and  $1.73 \pm 0.11$  Bq/kg, respectively. While, the average of AED (mSv/y) for vegetables and fruits samples was  $0.067 \pm 0.018$  and  $0.082 \pm 0.005$ , respectively. Also, the results showed that the average value of AED from fruits consumption is larger than in vegetables, but the result is not significant. All results of the  $C_{Ra}$  and AED of the studied samples had been compared with the worldwide reported value (median). Accordingly, it was found that all findings were lower than that of the recommended limits of the UNSCEAR 2000. Finally, based on present investigations, no health risk expected when considering eating vegetables and fruits of Al-Najaf of Iraq.

**Keywords:** Alpha emitters, Food contamination, Radium-226, Herbal samples, Al-Najaf.

## INTRODUCTION

It is customary that people are exposing to natural radiation in everyday life. The basic components that support human beings' continuous life is sought from the soil, water, air and vegetation. In this regard, some of the latter components are either inhaled or ingested into the body where they contain measurable amount of radioactivity (Abojassim et al., 2021). It should be noted that the specific metabolism of different plants types might lead to an accumulation of radio-nuclides that depends upon the physico-chemical characteristics of the soil (Engelbrecht, 2020). Fertilizers affect the increase in the concentrations of nuclides that occur naturally in the soil and therefore in plants at higher rates in agricultural areas (Ba et al., 2022; Najam et al., 2022).

Thus, the naturally occurring and man-made nuclides are transmitted to crops through the soil as one of the steps of the plant food chain through the roots, which is the initial step of this chain (Mukherjee, 2022). There may be an increase in the amount of risk that the human population might expose to via food chain (Shaw, 2018). Some of the aforementioned radiations are originated from natural sources, while others are from artificial sources. To illustrate, the natural sources of radiation include cosmic, terrestrial and internal radiations. By contrast, the artificial sources include medical radiation-based procedures, commercial radioactive materials-based products, and finally the fallout from nuclear testing (Abojassim et al., 2021). The major terrestrial elements of radioactivity include uranium, thorium, potassium and together with their corresponding decay products namely: 'radium and radon' (Shaw, 2018). Radium deposits in the bones up to 20%, causing necrosis and decomposition of the bones due to its radioactive activity that repels calcium from them. The danger of internal and external exposure to it lies in the alpha emitters that kill and mutate cells, and an isotope Radium-226 ( $^{226}Ra$ ) is the most toxic. The radium is a natural radioactive element. It is existed in the uranium and thorium series of the crust of the earth. In this context, the radium was used in many applications such as its use as a self-illuminating material in dial clocks, medical diagnoses and therapy (radiation therapy and brachytherapy) (Abojassim et al., 2021). The most abundant of the radium isotopes is  $^{226}Ra$  that is a natural arises as a decay product of uranium-238 ( $^{238}U$ ).  $^{226}Ra$  is an alkaline earth metal with physiological and environmental qualities similar to calcium and barium, making it an important radiotoxic radionuclide (L'annunziata, 2016).  $^{226}Ra$  being an alpha-emitter and having a half-life about 1620 years, it is of utmost interest because when deposited in internal organs of humans, it is known to

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cause severe radiation damage resulting from the alpha particles and short-lived daughter radionuclides of high specific activity emitted in its decay process (Zimmermann, 2007).

Such severe radiation damage may cause cancer as in the case of the early radium dial painters. Groundwater and Ra-bearing interact of components like rocks, soil, ore bodies, and other materials are natural sources of radium in groundwater then in some foods. Currently, chemical based agricultural fertilizers are key for the purpose of agricultural activity. They can help in increasing the production of crops and therefore improving the nutrient lands characteristics. Nevertheless, it is possible to encounter the negative impact of aforementioned fertilizers. For example, it includes the contamination of the agricultural lands by trace elements, and certain types of the naturally occurring radioactive materials (NORM) (Azeez et al., 2020).

Consequently, the latter argument can highlight that the usage of the fertilizers may involve certain types of hazards due to radiation to the farmers and then to general public (Abojassim, 2021). In this regard, the radium can result in some carcinogenic diseases to include lung, bone, skull, and nasal passage cancers when it is being inhaled. In case of internal exposure to large amounts of radiation, the radium has been reported to induce the cancer of bone cancer in animals as well as humans. In Iraq, animals become contaminants when they feed on radium-226 contaminated plants (vegetables and fruits) and water; those radionuclides are transferred to their bodies and products such as milk and dairy products, eggs, meat. As a result, contamination occurs primarily because of animals' ingestion of contaminants. The monitoring of the concentrations of radium in either sample (i.e., vegetables and fruits) had been the research focus for many published studies worldwide (Hashim and Najam, 2015; Girault et al., 2021; Hassan and Rashid, 2022).

This research was focused on investigating the radium-226's concentrations in samples of vegetables and fruits which are consumed in Najaf, Iraq utilizing the technique of nuclear track detectors (e.g., LR-115 type-II).

## MATERIALS AND METHODS

### Sample collection and preparation

Nine samples of vegetables together with nine samples of fruits were collected from the local markets of Najaf, Iraq as shown in Tables 1 and 2, respectively. After the collection of the required samples decided for this research from markets, they were cleaned and dried using electric oven at 60 Co for one day. Next, the samples were crushed via utilizing an electric mill. To reach the homogeneity of the samples, they were sieved using a 0.8 mm-pore-size sieve. Then, the resulted sample powder was placed in a plastic cup whose dimensions: 5 cm diameter, 7 cm length and 130 ml volume). The samples were labeled using a code and the country of origin. The corresponding net were determined and recorded utilizing digital balance with high accuracy. Finally, the plastic cups were stored for around one month before the counting process begins. This is in order to reach the secular equilibrium among the isotopes of natural decay series (Abojassim et al., 2016).

**Table 1 - Samples of vegetables under study.**

No.	Name of vegetables	Code	Country of origin
1	Celery	V1	Iraq
2	Parsley	V2	
3	Watercress	V3	
4	Onion	V4	Iran
5	Lettuce	V5	
6	Zucchini	V6	
7	Bell Pepper	V7	
8	Chili Pepper	V8	India
9	Garlic	V9	China

**Table 2 - Samples of fruits under study.**

No.	Name of fruits	Code	Country of origin
1	Rearrange	F1	Iraq
2	Pomegranate	F2	
3	Pear	F3	
4	Watermelon	F4	Iran
5	Apple	F5	
6	Banana	F6	Egypt
7	Orange	F7	
8	Lemon	F8	Turkey
9	Apricot	F9	

### Methods of measurement

After end time equilibrium, A piece of LR-115 type-II a sensitive cellulose nitrate (C<sub>6</sub>-H<sub>8</sub>O<sub>8</sub>-N<sub>2</sub>) detector with area 1x1 cm<sup>2</sup> and thickness of 12mm was placed at the bottom of each cylinder cover, with samples at the bottom of cylinder and then sealed for exposure of 90 days. Detectors were etched with sodium hydroxide (NaOH) solution of 2.5 (which is prepared by dissolving 40gm of NaOH in 0.4 Litter of distilled water) normality in temperature at 60 °C within 90min in order to show the tracks (Hady et al., 2016). The tracks count per unit area of the studied samples were counted using an optical microscope (Novel N-120A, 400X magnification power).

### Statistical analysis

In this work, the statistical analysis has been carried out by using the statistics software package SPSS version 23.0 for windows.

### Theoretical equations

Track density ( $\rho$ ) of each sample (track/cm<sup>2</sup>) was measured using equation (1) as follows (Abojassim, 2021):

$$\rho \left( \frac{\text{Track}}{c^2} \right) = \frac{\text{No.Tracks}}{\text{Area of view}} \quad (1)$$

The equation (2) was used to determine effective radium-226 content ( $C_{Ra}$ ) in samples, as following (Ibrahim et al., 2021; Hashim et al., 2021):

$$C_{Ra} (\text{Bq. kg}^{-1}) = \left( \frac{\rho}{KT_e} \right) \left( \frac{hA}{M} \right) \quad (2)$$

where, h: refers to the height of the sample inside the plastic cup (5 cm), A: refers to the area of plastic cup, M: refers to the mass of sample, K: represents the calibration factor which is equal to (0.256 tracks.cm<sup>-2</sup>. d<sup>-1</sup>/ Bq.m<sup>-3</sup>) and T<sub>eff</sub> represents the time of actual exposure which was calculated using equation (3) (Olewi et al., 2021):

$$T_{\text{eff}} = \left[ T - \lambda_{Rn}^{-1} (1 - e^{-\lambda_{Rn} T}) \right] \quad (3)$$

where  $\lambda_{Rn}$  represents the decay constant for <sup>222</sup>Rn.

Annual effective dose (AED) based on specific activity <sup>226</sup>Ra in unit Bq/kg ( $C_{Ra}$ ) were calculated using (4), as following (Abojassim and Lawi, 2018):

$$AED \left( \frac{\text{nSv}}{y} \right) = C_{Ra} \times I \times CF \quad (4)$$

where, I is consumption rate of samples which it is equal 60 kg/y for vegetables samples and 170 kg/y for fruits samples, and CF is conversion dose factor which it is equal 280 nSv/Bq (UNSCEAR, 2000).

## RESULTS AND DISCUSSIONS

The results of  $C_{Ra}$  in Bq/kg for vegetables samples in the present study as well as results of annual effective dose in mSv/y are given in Table 3. From Table 3, it is show that, the highest value of  $C_{Ra}$  was 10.291 Bq/kg in sample V4 (Onion, made in Iran), while the lowest value was 1.667 Bq/kg in sample V2 (Parsley, made in Iraq) with an average value of 3.98±1.08 Bq/kg. Also, from same Table 3, it is show that, the range values of annual effective dose due to <sup>226</sup>Ra concentrations were ranged from 0.028 mSv/y to 0.173 mSv/y, with an average value of 0.067±0.018 mSv/y. Table 4 illustrates the results of  $C_{Ra}$  in fruits samples of the present study. Table 4 show that the range of  $C_{Ra}$  in fruits samples in the present study varies from 1.232 Bq/kg in sample F5 (Apple, made in Iran) to 2.174 in sample F6 (Banana, made in Egypt), with an average value of 1.73±0.11 Bq/kg. The annual effective dose due to <sup>226</sup>Ra concentrations were determined for various samples of fruits which ranged from 0.059 mSv/y to 0.103 mSv/y with an average of 0.082±0.005 mSv/y. The results of <sup>226</sup>Ra concentrations in the collected vegetables and fruits samples under study were lower than the world median according to UNSCEAR 2000 (UNSCEAR, 2000) which is 32 Bq/kg. Also, this indicates that the annual effective dose in all vegetables and fruits samples was lower than the permissible limit of internal exposure due to ingestion which equal 0.3 mSv recommended by UNSCEAR 2000 (UNSCEAR, 2000). From results in Tables 3 and 4, it was found that there was a significant difference across all samples when considering the values of radium concentrations in vegetables and fruits samples due to geochemical composition and origin of soil cultivation kinds in this location which is taken of samples.

**Table 3 - Results of  $C_{Ra}$  and AED in vegetables**

No.	Code	$C_{Ra}$ (Bq/kg)	AED (mSv/y)
1	V1	1.884	0.032
2	V2	1.667	0.028
3	V3	2.029	0.034
4	V4	10.291	0.173
5	V5	3.189	0.054
6	V6	9.711	0.163
7	V7	2.319	0.039
8	V8	1.957	0.033
9	V9	2.826	0.047
Average ± S.E		3.98±1.08	0.067±0.018

$C_{Ra}$ = Effective radium-226 content; AED= Annual effective dose; V= vegetables sample.

**Table 4 - Results of  $C_{Ra}$  and AED in fruits.**

No.	Code	$C_{Ra}$ (Bq/kg)	AED (mSv/y)
1	F1	1.449	0.069
2	F2	1.739	0.083
3	F3	1.594	0.076
4	F4	2.029	0.097
5	F5	1.232	0.059
6	F6	2.174	0.103
7	F7	2.102	0.100
8	F8	1.305	0.062
9	F9	1.957	0.093
Average ± S.E		1.73±0.11	0.082±0.005

$C_{Ra}$ = Effective radium-226 content; AED= Annual effective dose; F= fruits sample.

The comparison of the average value of  $C_{Ra}$  and AED between vegetables and fruits samples in the current research can be seen in Figures 1 and 2, respectively. There are two ways to transfer natural radionuclides to plants and one of these two methods of natural radionuclides transmitted method of absorption indirectly from the soil via the roots. While the crops are growing up in a given contaminated soil, the radioactivity is driven from the soil to the plant roots and from there into the plant stem, and then into leaves of the fruit tree (Hussain and Rani, 2010). That is why the emergence of radium concentrations in vegetables are higher than in the fruit (Figure 1), because most vegetables have roots and

leaves while the fruits have few roots. Regarding the annual effective doses, it had been found that for the fruits the value was larger than the dose in vegetables (Figure 2). This larger value for fruits is due to the high consumption rate of samples by adult. Finally, we can say the vegetables and fruits in the present study are no health hazards due to radium-226 concentrations.

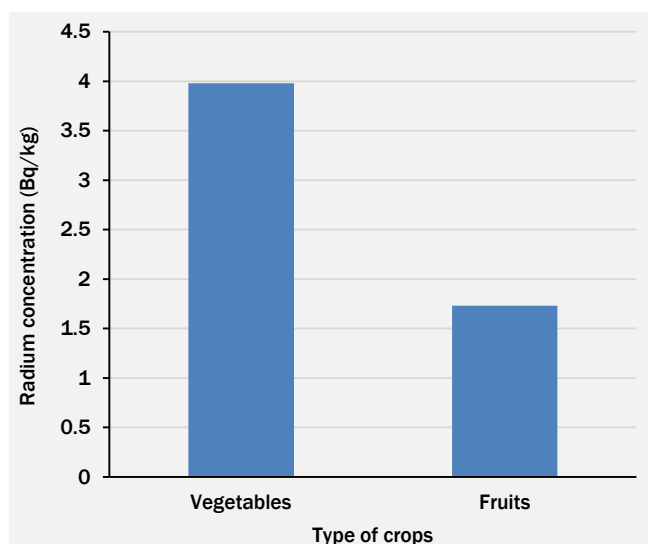


Figure 1 - Comparing of the average value of  $^{226}\text{Ra}$  concentrations between vegetables and fruits samples in the present study.

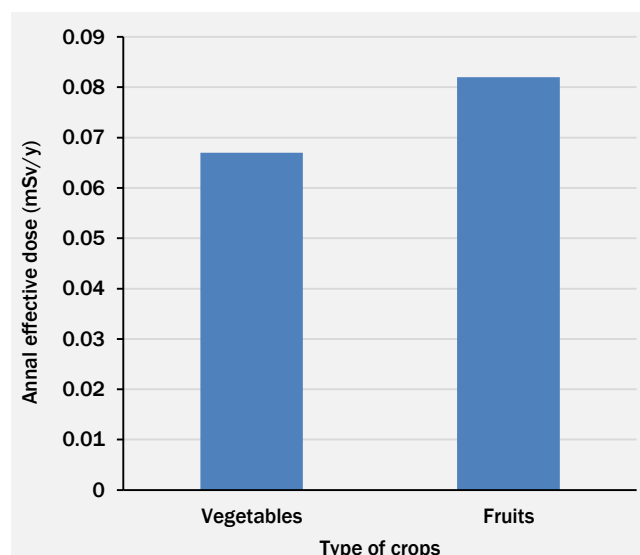


Figure 2 - Comparing of the average of AED between vegetables and fruits samples in the present study.

## CONCLUSION

According to the findings obtained by this research, one can conclude that the radium-226 concentrations values together with annual effective dose for eighteen samples of vegetables and fruits were noticed to be below the worldwide median value (i.e., 32 Bq/kg and 0.3 mSv/y) which were reported by UNSCEAR 2000. It was also found that the average concentration value of  $^{226}\text{Ra}$  in vegetables samples was seen to be larger than in the fruit's samples. Concerning the annual effective dose resulted from the fruits samples was noticed to be larger than in vegetables samples. The findings of this work, however, demonstrate that vegetables and fruits (local and imported) in the Iraqi market in Najaf governorate consumption does not pose any severe health risks to humans and animal. Animals receive radium-226 through their food (such as vegetables and fruits) and water. Therefore, we must recommend the necessity of detection on the contamination occurs primarily as a result of animals' ingestion of this food.

## DECLARATIONS

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### Authors' contribution

All authors are contributed to the present work.

### Conflict of interests

There is no conflict of interests.

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### Data availability statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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