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Quantitative analysis of natural radioactivity levels in legumes and grains: Exploring implications for human health

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► ***Original article***

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ABSTRACT

Background: Natural radioactive nuclei in the earth's geology are being increasingly studied worldwide due to their potential public health implications. This research is crucial, as legumes, which are a primary food source for many people, commonly contain such radionuclides. The study aimed to evaluate the background radiation in legumes and grains collected from an Iraqi market using two techniques: a scintillation detector system and a CN-85 detector. ***Materials and Methods:*** As discussed in the first part of this work, after samples were collected, they were dried at 70 °C, ground up, weighed, and stored in Marlin beakers for four weeks to reach equilibrium before ^{222}Rn and ^{226}Ra. NaI (Tl) scintillation detectors were used. The samples were then sealed in test tubes for four weeks to ensure further balance. Detectors were used to measure alpha release for 79 days before the samples were treated with 2.5N NaOH at 60 °C for 3 hours. All samples were then placed in a water bath, washed with distilled water, and dried, and the nuclear detector effects were calculated using microscopy. ***Results:*** The ^{238}U emissions ranged from 5.86±0.15 to 1.3±0.12 Bq/kg, while the 232Th was between 3.82±0.14 and1.1±0.11 Bq/kg; ^{40}K was between 59.53±2.21and 414.16±3.81 Bq/kg. The Ca in the airspace between the samples and the detector ranged between 288.766 and 656.643 Bq/m^3, while the CRn in the samples ranged between 4,827.885 and 10978.440 Bq/m^3. ***Conclusion:*** The results closely aligned with or slightly exceeded those of previous studies. All results fell within UNSCEAR 2000 internationally permitted limits, however, suggesting that such produce offers no danger to human life or health from radioactivity.

INTRODUCTION

Radioactive isotopes occur naturally in the environment, with their origins in both terrestrial sources and cosmic radiation. These elements are commonly incorporated into biological systems through metabolism by plants and animals, thus exposing humans at all stages of the food chain [1]. The uptake of radioactive elements by plants and animals is primarily controlled by the chemical properties of such elements rather than their radiological properties, however. For example, plants uptake calcium (Ca) and potassium (K) from soil through their root systems without distinguishing between stable isotopes (e.g., ^{40}K) and radioisotopes (e.g., ^{45}Ca) due to their identical biochemical pathways [2].

Agricultural fertilizers offer another source of environmental radiation. Phosphate fertilizers, derived primarily from sedimentary phosphate rocks [3] , may contain concentrations of uranium ranging from 0.5% to 20 × 10^{-2}%, for example. During the decay of uranium, Radon (^{226}Ra) is formed at significant rates, and this is transported mainly in phosphogypsum, a by-product with high solubility in sulphates [4].

Further, controlled radiation exposure provides viable alternatives to conventional chemical disinfection for pest control in many food commodities, including cereals, pulses, spices, fruits, and vegetables. This method not only inhibits the growth of microbes (including pathogens harmful to humans) but also prevents the germination and enzymatic degradation of plant cells, thereby enhancing food safety and extending shelf life [5].

Terrestrial radioisotopes, particularly thorium (Th), uranium (U), and potassium (K), thus enter the human body primarily through dietary consumption and thus constitute important sources of internal and external radiation exposure. Inhalation of these isotopes, being less common, is purely a secondary route of exposure [6]. A critical priority for those concerned with human health is thus to elucidate the behaviour of natural radionuclides across various environmental and biological matrices, using the resulting data to serve as basic criteria for robust radiological risk assessments [7].

Long-term exposure to alpha radiation emitted by radionuclides such as radon gas is associated with severe pathological effects, including functional

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impairments in the respiratory system and elevated risk of lung cancer [8]. Alpha particles, characterised by high linear energy transfer and rapid attenuation in matter, induce significant damage to biological tissues. Trace quantities of alpha-emitting isotopes such as uranium and thorium and their decay products can infiltrate living organisms through both dietary and environmental pathways, potentially posing a persistent health hazard [9]. Certain food commodities contain low but measurable levels of naturally occurring alpha- and gamma-emitting radionuclides, which may compound any risks due to the combined radiological and chemical toxicity of the resulting isotopes. Chronic exposure to such emissions can thus induce cumulative damage to various organ systems, particularly in terms of DNA strand breaks and oxidative stress [10].

These risks underscore the critical need to quantify and monitor radioactivity in widely consumed foodstuffs. This study therefore addresses this imperative by evaluating natural radioactivity levels in ten varieties of legumes and grains commercially available in Iraq. By integrating two complementary analytical approaches, namely a gamma-ray scintillation detector for quantifying ^{238}U, ^{232}Th, and ^{40}K activity concentrations (including derived hazard indices, absorbed dose *[AD]*, and radium equivalent activity $[Ra_{eq}]$, and a CR-39 nuclear track detector (CN-85) for assessing radon concentrations, effective radium content, radon exhalation rates, and uranium levels, this work provides a comprehensive radiological profile of these staple food items.

The findings thus hold direct relevance for the shaping of food safety regulations and public health policies. Chronic dietary intake of radioactive elements, even at low concentrations, may contribute to both carcinogenic and non-carcinogenic health burdens due to the bioaccumulation of radionuclides. By establishing baseline data on natural radioactivity in legumes and grains, this study can inform risk assessment frameworks and support the development of strategies to mitigate population-level exposure in regions reliant on such dietary staples if necessary.

MATERIALS AND METHODS

Materials

Ten food samples were collected from the local markets in Iraq, as shown in table 1. These were dried in an oven, a Toshiba model TL2-SAC25GZE-GR manufactured in China, at about 70 °C for 3h to get remove all moisture before being ground in a grinder (Silver Crest, China). After that, the samples were screened using a 500 µm locally-made sieve before being weighed on a 1byOne Digital Scale, made in China). A 500 gm sample was taken of each foodstuff, and these were stored in marlin beakers for about four weeks in order to achieve equilibrium between ^{222}Rn and ^{226}Ra [11]. An NaI (TI) scintillation detector from BICRON with crystal dimensions of 1.5×2 inch, manufactured in Germany, was used with the CASSY lab program (1,024 channel MCA). The detector was energy-calibrated using a gamma-ray radioactive source ^{137}Cs, again sourced from a German company, Phywe, while ^{22}N, and ^{60}Co were sourced from an LD company. For the resolution of 7.5% energy at 662 keV of ^{137}Cs, the counting time was set at 24 hours in order to obtain the best peaks in the recorded spectrum.

Table 1. The details of the samples used in this research.

No.	Sample name	Name of Company	Country of manufacture
1	Lentils (*Lens culinaris*)	zer	Türkiye
2	Fava bean (*Vicia faba*)	Altunsa	Türkiye
3	Peas (*Pisum sativum*)	zer	Türkiye
4	Bean (*Phaseolus vulgaris*)	Altunsa	Türkiye
5	Wheat (*Triticum aestivum*)	Al mahaba	Iraq
6	Chickpeas (*Cicer arietinum*)	zer	Türkiye
7	Cowpea (*Vigna unguiculata*)	Altunsa	Türkiye
8	Rice (*Oryza sativa*)	Kohinoor	India
9	Barley (*Hordeum vulgare*)	Babil duy	Türkiye
10	Corn (*Zea mays*)	Al-Nema	Iraq

Roughly 16 grams of each sample of grain or legume were stored in each cylindrical tube; the tubes had a height of 6.5 cm and a diameter of 4 cm and were numbered appropriately. CN-85 Solid State Nuclear Tracking Detectors, manufactured by Kodak-Pathe in France [12-14] with a thickness of 12 µm (known as SSNTD) were used, with dimensions 1 cm × 1 cm. These trigger a certain number on the top right corner of the detector to facilitate the process of gathering information and distinguishing between the detectors for various samples, and they were attached to each detector on the inner surface of the top of the tube using two-sided adhesive tape. A sensitive balance, with a sensitivity of 10^{-3}, (type VIC-303 US) was also fitted to measure the weights of samples (Accul AB Sartorius, Germany).

In the second section of the experiment, the samples were deposited in test tubes based on the sealed can method, as shown in Figure 2. The samples were held for four weeks to bring the radium and radon components of the decay sequence into balance; detectors used to assess the nuclear impact of food samples were then utilised for 79 days to measure any alpha radiation released by the samples. After this exposure, samples were put in a 2.5N NaOH solution for three hours at a temperature of 60±1 °C. All detectors were put in a water bath (Memmert WNB22, Germany). This method allowed a display of the tracks of alpha particles as they collided with each detector and its derivatives. After that, the detectors were removed from the water bath and then washed with distilled water. Once the detectors were dried, the effects of any nuclear impacts were counted using an optical microscope from Olympus

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BX53 (Japan), with a magnification of 400x; this is typically used to ensure precise and accurate analysis of such tracks.

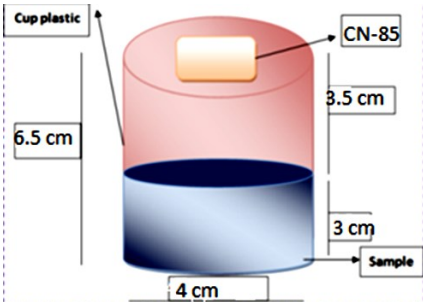

Figure 2. Sealed can technique.

Methods

Gross alpha and gross gamma radioactivity measurements were performed using Gamma spectroscopy and the Sealed Can Technique, which uses CN-85 solid state comparators. To measure the gamma radioactivity, an NaI(Ti) detector was used to calculate the following parameters:

Specific activity (A)

The specific activity of each radioactive nucleus was calculated in the samples studied using equation 1 [15]:

$$A(B_q \times Kg^{-1}) = \frac{(N-B)}{t \times \varepsilon \times I_\gamma \times m} \quad (1)$$

Where; A is specific Activity, B is background count, N is total count (sample+ Background), t is time (sec), I_γ is the intensity of specific energy, ε is the gamma-ray efficiency of the detector, and m is the mass of the sample in kg.

Minimum Detectable Activity (MDA) is a fundamental term in the realm of radiological measurements and nuclear research. The term refers to the minimum detectable level of radioactivity that a particular type of measuring equipment can accurately identify. Several factors, such as the intensity of background radiation, the efficacy of the detection system, the counting interval, and the required statistical confidence level, influence the MDA for a given method, and this was thus calculated using equation 2 [16].

$$MDA = \frac{4.66\sqrt{B}}{\varepsilon \times m \times t} \quad (2)$$

The resulting factor, 4.66, corresponds to a confidence level of 95%.

External hazard index (H_{ex})

The external hazard index (H_{ex}) guides assessment of the risk of gamma radiation. This is calculated using equation 3 [17]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (3)$$

Where; A_{Ra}, A_{Th}, and A_k represent specific activity in the radium series, thorium series, and potassium series, respectively [18]. The H_{ex} parameter should be less than or equal to one; if it is larger, this indicates a radiation hazard.

Radium Equivalent (Ra_{eq})

The radium equivalent, Ra_{eq}, can be calculated using equation 4:

$$Ra_{eq}\left(\frac{Bq}{Kg}\right) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (4)$$

Absorbed dose rate in air (AD)

The specific activity of the chains uranium, thorium, and potassium was used to find the absorbed dose (AD); the unit is (nGy h^{-1}) [18].

$$AD = 0.92A_U + 1.1A_{Th} + 0.08A_K \quad (5)$$

In this case, AD = 0 where 0.92, 1.1, and 0.080 $nGyh^{-1}$/ Bq kg^{-1} are used as the conversion factors for ^{238}U, ^{232}Th, and ^{40}K, respectively.

Radon concentration

To calculate ^{222}Rn concentration levels in the different samples of food, the radon activity density, C_a , in the air above the samples in each was determined using equation 6 [19]:

$$Ca\ (Bq/m^3) = \rho\ k/t \quad (6)$$

Where; C_a is the ^{222}Rn concentration in the sample (Bq/m^3), ρ is the surface density of tracks on the exposed detectors ($Track/cm^2$), t is the exposure time (79 days), and k is the calibration factor, which was determined experimentally to be 0.256 $track.cm^{-2}$/ $Bq.d.m^{-3}$ [20].

The dissolved radon concentration, C_{Rn} in rice was calculated using equation 7 [21, 22]:

$$C_{Rn} = \frac{C_a \lambda h T}{L} \quad (7)$$

Where C_a is the radon concentration in ambient air (Bq/m^3), λ is the decay constant for radon (d^{-1}), h is the distance from the surface of rice to the detector (m), T is the time of exposure, and L is the depth of the sample (m).

Effective radium content

Over a four-week period, effective equilibrium for the radium-radon components of the decay series was attained (around 98% efficacy). Radon alpha analysis was used to determine the steady-state activity concentration of radium once the radioactive equilibrium was attained, as the activity concentration of radon begins to increase proportionate to time T after the can is closed. To find the effective radium content of the samples, equation 8 was used [23, 24]:

$$C_{Ra} = \frac{\rho h A}{K T_e M} \quad (8)$$

Where T_e is the effective exposure time, related to the actual exposure time T, and the decay constant λ is used for ^{222}Rn [25].

$$T_e = T - \frac{1}{\lambda}(1 - e^{-\lambda T}) \quad (9)$$

Radon exhalation rate

The radon exhalation rate in terms of area was

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calculated using equation 10 [26]:

$$E_A = \frac{CV\lambda}{AT_e} \quad (10)$$

Where; E_A is the radon exhalation rate (Bq/m^2. H), C represents the integrated radon exposure (Bq.m^{-3}.h); V is the effective volume of the can (4.3295×10^5 m^3), T_e is the exposure time (79 days), λ is the decay constant for radon, and A is the area of the tube (9.6211×10^{-4}m^2).

The radon exhalation rate in terms of mass was then calculated using equation 11 [26]:

$$E_M = \frac{CV\lambda}{MT_e} \quad (11)$$

Here, E_M is the radon exhalation rate in terms of mass (Bq/ kg.h) and M is the mass of the sample.

Uranium concentrations

To calculate the uranium concentration of edible oil samples, C_U can be defined as the ratio between the uranium weight in the sample (W_U) and the sample weight (W_S) according to the equation 12, as measured in part per million (ppm) units [27]:

$$C_U(ppm) = \frac{W_U}{W_S} \quad (12)$$

To measure environmental radioactivity, the Becquerel (Bq) per unit mass ration was used to express the amount of Uranium present. The following conversion factors (equation 13), from concentration unit to activity unit in Bq.kg^{-1}, were used, as provided by the International Atomic Energy Agency (IAEA) [28, 29]:

1ppm of U = 12.35 Bq.kg^{-1} of ^{238}U (13)

Statistical analysis

IBM SPSS Statistics Software version 25 (IBM corporation Chicago, USA) was used in the calculations for this research. This included the analysis of all resulting data, and the calculation of the relevant MDA. Significance levels were set at the 95% confidence interval level (Alpha=0.05).

RESULTS

Ten different samples of foodstuffs were selected from local Iraqi markets. Gamma spectroscopy using an NaI (TI) detector and the sealed can technique (Somogyi, 1990) [30] containing nuclear track detectors of type CN-85 were used to measure the natural radioactivity of gamma and alpha emitters, respectively.

Specific activity (A) analysis

Table 2 shows the effective concentrations of the uranium, thorium series and potassium elements, showing that the values of the uranium series ranged from 5.86±0.15 to 1.3±0.12 Bq/kg in (Lentils) and (Fava bean) respectively, with an average of 3.483±0.135 Bq/kg. The thorium series was between 3.82±0.14 and 1.1±0.11 Bq/kg in (Barley) and (Rice), while the minimum value of potassium (59.53±2.21 Bq/kg) occurred in 0 (Corn) and the maximum value (414.16±3.81 Bq/kg) occurred in (Beans), giving an average rate of 260.678±3.175 Bq/kg. Table 3 shows the extent of convergence of the results of the current study with similar local and international studies. The results are lower than the internationally permissible limits, which are much lower than the recommended limits (45, 32, and 412 Bq^{-1} for uranium ^{238}U, ^{232}Th, and ^{40}K, respectively) [31].

Table 2. Specific activity for uranium chine (^{238}U), thorium chine (^{232}Th), and potassium(^{40}K).

Sample name	Specific activity (**Bq/kg**)		
	^{238}U Bq/kg	**^{232}Th Bq/kg**	**^{40}K Bq/kg**
Lentils (*Lens culinaris*)	5.86±0.15	1.37±0.11	269.84±3.25
Fava bean (*Vicia faba*)	1.3±0.12	2.42±0.13	319.18±3.45
Peas (*Pisum sativum*)	4.72±0.14	1.5±0.11	223.5±3.05
Bean (*Phaseolus vulgaris*)	4.51±0.14	1.3±0.11	414.16±3.81
Wheat (*Triticum aestivum*)	4.84±0.14	1.39±0.11	133.17±2.62
Chickpeas (*Cicer arietinum*)	3.59±0.14	1.4±0.11	296.79±3.36
Cowpea (*Vigna unguiculata*)	3.66±0.14	2.28±0.12	355.58±3.59
Rice (*Oryza sativa*)	2.71±0.13	1.1±0.11	383.84±3.7
Barley (*Hordeum vulgare*)	2.28±0.13	3.82±0.14	151.19±2.71
Corn (*Zea mays*)	1.36±0.12	1.25±0.11	59.53±2.21
Max	**5.86±0.15**	**3.82±0.14**	**414.16±3.81**
Min	**1.3±0.12**	**1.1±0.11**	**59.53±2.21**
Mean ± SD	**3.483±0.135**	**1.783±0.116**	**260.678±3.175**

*SD=Standard Divation

Table 3. Radionuclide concentration (current study) compared to previous similar local and international research.

No.	^{238}U	^{232}Th	^{40}K	country	Ref.
1	7.475	3.79941	385.2477	Iraq	[32]
2	5.35	2.20	178.17	ALBANIA	[33]
3	11.18	6.50	211.12	Nigeria	[34]
4	6.119	4.763	135.595	Iraq	[35]
5	4.04	2.34	146.61	Saudi Arabia	[36]
6	3.483	1.783	260.678	Iraq	The current study

External hazard index (H_{ex}), radium equivalent (Ra_{eq}), and absorbed dose rate in air (AD) analysis

Table 4 shows the values of the hazard index, absorbed dose rate in air (AD) and radium equivalent activity. The value of the hazard index varies between (Beans) and 0(Corn), with gave values of 0.1 and 0.02, respectively for an average rate value of 0.071, well within the acceptable range.

The absorbed dose rate in air ranged from 38.71 nGy/h to 7.39 nGy/h in (Beans) and 0 (Corn), respectively, for an average of 26.02 nGy/h, Finally, the Radium Equivalent Activity was between 38.26 Bq/Kg in (Beans) and 7.73 Bq/Kg in 0(Corn), for an average of 26.104 Bq/Kg.

Radon concentrations, effective radium content, radon exhalation rate, and uranium concentrations analysis

In terms of the sealed can technique, Table 5

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shows the density of the nuclear tracks formed on the surface of the CN-85 detectors, alongside the relevant radon concentrations, values of effective radium content, radon exhalation rate, and uranium concentrations in ten food samples. From table 5, the highest values were recorded in sample 0(Corn), and the lowest values were recorded in sample (Cowpea). The radon concentrations in the air spaces between the samples and the detectors ranged between 288.766 Bq/m^3 to 656.643 Bq/m^3, with an average of 479.430 Bq/m^3, while the radon concentrations in the samples ranged between 4827.885 Bq/m^3 and 10978.440 Bq/m^3, for a mean of 8015.606 Bq/m^3. Mass and surface measurements for Radon exhalation rates ranged from 6.450 mBq/kg.h to 14.667 mBq/kg.h and from 82.121 mBq/m^2.h to 186.740 mBq/m^2.h, respectively, with rates of 10.708 mBq/kg.h and 136.343 mBq/m^2.h on average.

Table 4. Hazard index(Hex), Absorbed Dose Rate in Air (AD), and Radium Equivalent Activity (Raeq).

Sample name	H_{ex}	Absorbed Dose Rate in Air (AD) (nGy/h)	Radium Equivalent Activity (Ra_{eq}) (Bq/Kg)
Lentils (*Lens culinaris*)	0.08	28.49	28.6
Fava bean (*Vicia faba*)	0.08	29.39	29.34
Peas (*Pisum sativum*)	0.07	23.87	24.07
Bean (*Phaseolus vulgaris*)	0.1	38.71	38.26
Wheat (*Triticum aestivum*)	0.05	16.64	17.08
Chickpeas (*Cicer arietinum*)	0.08	28.59	28.44
Cowpea (*Vigna unguiculata*)	0.09	34.32	34.3
Rice (*Oryza sativa*)	0.09	34.41	33.84
Barley (*Hordeum vulgare*)	0.05	18.39	19.38
Corn (*Zea mays*)	0.02	7.39	7.73
Max	0.1	38.71	38.26
Min	0.02	7.39	7.73
Average	0.071	26.02	26.104

Table 5. Track density (ρ), Activity concentration of radon (C_a), The concentration of radon gas in air (C_{Rn}), Concentration of radium (C_{Ra}), Exhalation rate of radon from a surface per unit area(E_M), Exhalation rate of radon from a surface per unit area(EA) and Concentration of uranium(CU) in different food samples of Iraqi market.

Sample name	(ρ) Track/cm2	(C_a) Bq/m^3	(C_{Rn}) Bq/m^3	(C_{Ra}) Bq/kg	(E_M) mBq/kg.h	(E_A) mBq/m^2.h	(CU) Bq/kg
Lentils (*Lens culinaris*)	9840.009	486.551	8134.664	1.438	10.867	138.368	18.771
Fava bean (*Vicia faba*)	7200.005	356.013	5952.191	1.052	7.952	101.245	13.735
Peas (*Pisum sativum*)	11439.980	565.664	9457.352	1.672	12.634	160.867	21.824
Bean (*Phaseolus vulgaris*)	7440.006	367.880	6150.598	1.087	8.217	104.620	14.193
Wheat (*Triticum aestivum*)	10799.990	534.019	8928.276	1.578	11.928	151.868	20.603
Chickpeas (*Cicer arietinum*)	12239.970	605.220	10118.700	1.788	13.518	172.116	23.350
Cowpea (*Vigna unguiculata*)	5840.000	288.766	4827.885	.853	6.450	82.121	11.141
Rice (*Oryza sativa*)	7600.006	375.791	6282.870	1.111	8.394	106.870	14.498
Barley (*Hordeum vulgare*)	11279.990	557.753	9325.083	1.648	12.458	158.617	21.518
Corn (*Zea mays*)	13279.960	656.643	10978.44	1.940	14.667	186.740	25.334
Mean	9695.992	479.43	8015.606	1.4167	10.708	136.343	18.496
Max.	13279.960	656.643	10978.440	1.940	14.667	186.740	25.334
Min.	5840.000	288.766	4827.885	.853	6.450	82.121	11.141

DISCUSSION

The background radiation in the legume samples used was found to be well within expected natural levels, we note that the concentrations of uranium, thorium chains and potassium for this study are lower than the results of the comparative study in table 3. It was also observed that the values of effective radium content and uranium concentrations ranged from 0.853 Bq/kg to 1.940 Bq/kg and from 11.141 Bq/kg to 25.334 Bq/kg, giving averages of 1.416 Bq/kg and 18.496 Bq/kg, respectively. The results of this study thus offer good convergence with the results of previous studies including various samples of rice that studied by Hashim *et al*. to various brands of Rice in Iraqi market [37] and the study of water in North Guilan province for Abbasi *et al*. [38], though they were higher than the results of some studies including study of Kurnaz *et al*. to water and dam water for Kastamonu city Centre-Turkey [39], Nevertheless, the results suggest that none of the tested foods

The radiation doses calculated for the consumption of these legumes were thus negligible and do not pose any significant health risk. These results are consistent with previous studies on the radioactivity of agricultural products, supporting the conclusion that natural radiation in legumes does not pose a health problem for humans under normal dietary consumption conditions. While the results of the study are reassuring, further research involving a larger sample size and a wider geographical scope would provide more comprehensive confirmatory data. In addition, investigating the effects of specific soil conditions and agricultural practices on the uptake of radionuclides would enhance understanding in this area. Overall, the levels of natural radioactivity in the studied legumes were normal and thus do not pose any significant radiological risk to human health.

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CONCLUSION

The study measured the natural radioactivity of gamma and alpha emitters using NaI(Tl) and CN-85 detectors. Variations in radioactive concentrations were observed across different samples, influenced by soil type, geological factors, and nuclear abundance. The findings were consistent with or only slightly higher than previous local studies, and all results remained within the international safety limits set by the United Nations Scientific Committee on the Effects of Atomic Radiation. This indicates that there is no health risk from radiation when the foods in these samples are consumed.

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