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Natural and Anthropogenic Radionuclides Concentration and Risk Assessment in Legumes and Grains, which Provides in a Basket Foodstuffs in Iraq Country

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Abstract

The present work studied the amount of natural and anthropogenic radioactivity in 13 different imported legumes and grain specimens of various origins, which are part of the basket of foodstuffs provided by the government to the Iraqi people. These specimens were analyzed using Canberra's sodium iodide NaI (TI) detector gamma spectrometer systems. The data gained revealed that the activity amounts (Bq/kg) for 214Bi, 137Cs, 40K (238U series), and 228Ac (232Th series) in all selected specimens were measured. Importantly, these investigated magnitudes are well within the international standards for radioactivity in foodstuffs, ensuring the safety of the food consumed by the Iraqi people. The mean radium equivalent activity (Raeq) in each specimen has been consistently below the 370 Bq/kg threshold. Each magnitude regarding indoor and outdoor yearly effective dosage corresponding for all specimens has been significantly lower than the international typical magnitudes (0.45 mSv/y concerning indoor as well as 0.07 mSv/y regarding outdoor). The subsequent radiological effects illustrate that the mean magnitude of annual total air absorbed rate dosage in these specimens is well below the limit of (1.0 mSv/y) based on the International Commission on Radiological Protection (ICRP).

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Introduction

Natural sources of ionizing radiation possess the inherent characteristic of emitting nuclear particles or Gamma rays (Al-Khafaji & Falah, 2020). Consequently, human beings are inevitably subjected to radiation from these sources. The primordial radionuclides comprise the natural series, including ^{238}U, ^{232}Th, and the nonseries ^{40}K. These radionuclides are often characterized by their lengthy lifespan, with a half-life exceeding one hundred million years ago (Tholkappian et al., 2018; Alharbi & El-Taher, 2013; Al-Khafaji & Falah, 2020; Al-Khafaji et al., 2023). Genuinely occurring radioactive substances, known as natural radionuclides, may be found in several elements of the human environment, including earth material, water, air, foods, and even our bodies. The primary sources of ionizing radiation from natural radioactivity are the long-lasting isotopes ^{238}U and ^{232}Th and their respective decay series, as well as ^{40}K (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 2017). Cement factories also include radioactive materials like uranium and thorium in their environment (Anthes & Anthes, 2020).

Examining these radioactive isotopes in food is crucial to the environmental surveillance program. These naturally occurring radioactive sources are the primary cause of the radiation dosage that humankind is exposed to(Ashok et al., 2019; Gunasekaran et al., 2023). Radioactive pollution, often known as radiological pollution, refers to the unintentional or undesired existence of radioactive chemicals on surfaces or inside gases, liquids, or solids (which involve the human being) (Brigden et al., 2002). Pollution might impact individuals, locations, animals, or items like clothes (Bureau, 2018; Canton, 2021). After an atmospheric nuclear weapon detonation or a breach in nuclear reactor containment, the animals, plants, people, soil, and air in the surrounding area will be contaminated by radioactive fuel and fission products (Al-Hamzawi, 2017; Charles, 2001). Food may get contaminated with radioactive elements due to an ambiguous emergency. Radioactive elements may contaminate the food's surface such as animal feed, vegetables, or fruits when exposed to radioactive substances in the air or via rainfall (Brynjolfsson, 2002; Cifuentes et al., 2023). Once radioactive compounds in the plume settle on water, soil, or plants, they have the potential to infiltrate the food chain. Initially, animals and plants may be exposed to radiation on their surfaces.

Additionally, they may experience tissue pollution through inhalation or ingesting radioactive substances in their food. It is worth noting that animals that consume a significant amount of contaminated items can accumulate radioactivity in their tissues. The level of tissue pollution will decrease gradually over time (Hussain & Al-Khafaji, 2020; Hussain & Al-Khafaji, 2020). Radionuclides may penetrate the human being via ingestion and inhalation. The radionuclides that are consumed could gather in restricted areas of the body. Due to consuming radionuclides through food intake could contribute significantly to the radiation dosages received by different body organs. This pathway is also essential to consider for long-term health implications.

Pathways of Radionuclides through the Environment

Radionuclides disperse through the environment via the same routes as other substances. They propagate via the atmosphere, traverse various aquatic environments (including surface water and subterranean), and permeate the food sites. Radionuclides might be introduced into the human being by ingestion (consumption

of food or drink), inhalation, or dermal absorption. The following passage delineates how radionuclides traverse the environment and enter the human being (Hussain & Al-Khafaji, 2022). There are several routes by which radionuclides may travel through the environment (Al-Masri et al., 2004).

- Atmospheric pathway
- Water pathway
- Food chain pathway

Contamination of Grain, Legumes and Rice

Following the rice, legumes, grain, and harvest are susceptible to pollution only throughout storage, with only the exterior layers being affected. Once radioactive fallout happens throughout growth, radionuclides will be transported into the rice, legumes, and grain via plant development. Choosing representative rice, legumes, and grain samples throughout the harvest season is straightforward. Once the nuclear fallout happens in winter, root absorption will only poison the grain in the next growth season (Hussein et al., 2018).

Levels of radionuclides in plants vary typically from a few tens of Becquerel (Bq) to the several hundred of Becquerel per kilogram (International Atomic Energy Agency, 2007). The radionuclides that exist in the fertilizers s in winter, root absorption will only poison the grain in the next growth season are uranium and thorium decay series as well as potassium. Besides, the concentration of depending on the origin of the components. Measurement and assessment of natural radioactivity is necessary because in most countries of the world, the study of naturally occurring radiation and environmental radioactivity was carried out (Murray, 1989; Weiss & Shannoun, 2018). During the past decades, the agricultural activities in environment widely grew up, especially fertilizers, pesticides, and some other chemicals to improve soil properties, enhance the quality of the crop products and to get more gain in terms of crop quantity as well. In other words, their concentration could be increased as contaminants over the time (Jibiri et al., 2007). From many countries, to establish a baseline data to the natural radioactivity levels, measurement of natural radionuclides in environmental elements has been carried out (Kazempoor et al., 2022).

The objective of this examination is not only to identify the quantities of natural radioactivity in 13 specimens of legumes and grains gained from various sources and distributed by the Iraqi government as part of the basket commodities in 2022 but also to compute the radium equivalent activities yearly dosage equivalent, internal and exterior indices of hazard, extent index, and the gamma dosage rate utilizing a gamma spectrometry technique using a thallium-doped sodium iodide detector (NaI(Tl)) has two principal advantages:

- It can be produced in large crystals, yielding good efficiency, and
- it produces intense bursts of light compared to other spectroscopic scintillators.

NaI(Tl) is also convenient to use, making it popular for field applications such as the identification of unknown materials for law enforcement purposes (Al-Khafaji & Falah, 2020; Knežević & Knežević, 2019).

Subsequently, it assessed the level of radioactivity and its impact on human well-being. This research is of utmost importance for the radiation safety field as it focuses on the presence of natural radioactive elements in food consumed by people, providing evidence-based data on the potential impact of radioactivity in food on human health (Jacquemain et al.,2014).

Material and Methods

Preparation and Collection of Specimens

In this investigation, specimens are selected from the basket of foodstuffs provided by the government to the Iraqi people in 2022. These specimens are coded as illustrated in Table (1), put in plastic bags, and stored in the lab for 30 days to obtain balance and suitable heat. Before counting, grinding the specimens to make a powder, then put the specimens in an oven (100 °C) for one hour to remove the moisture inside the mater (Yang, 2024)

Table 1. Grain and legume specimens

Specimen No.	Description	Origin
01 W	White Beans	Egypt
02 R	Rice	India
03W	White beans	Kirgizstan
04 B	Broken chickpeas	Kirgizstan
05 W	White beans	Argentina
06 W	White beans	turkey
07 B	Broken chickpeas	Bulgaria
08 R	Rice	Uruguay
09 B	Broken chickpeas	Russia
10 L	Lentils	Canada
11 R	Rice	USA
12 R	Rice	Thailand
13 F	The government provides flour	Iraq

The measurements are conducted using gamma spectrometry equipment equipped with Canberra's NaI (TI) detectors. The detector was encased in a lead shield that was 5 cm thick to minimize background interference. Additionally, a 0.3 cm copper layer was added to absorb x-rays released by the lead shield. The detector is calibrated for energy utilizing a set of reference gamma sources manufactured in the same geometry (Marinelli beaker) as the studied specimens. The efficiency calibration was conducted, and the curves were derived by fitting the experimental efficiencies for every specimen density. Efficiency curves are adjusted to account for the reduction in intensity and absorption of energy. The detectors' linearity was verified utilizing a mixed source provided by the Czech Metrology Institute for quality assurance. Certified foodstuff reference materials have been used in the specimens' counting geometry. The background spectrum is often gained using the same experimental circumstances as the measurements of the specimen and is utilized to adjust the estimated specimen activity. Each specimen is counted for an average duration of 3600 seconds to provide robust statistical significance (Knoll, 2010). The spectra are processed offline utilizing the Genie 2000 software from Canberra, which includes modules for peak search, nuclide identification, and activity and uncertainty computation. The activity of certain specimens is measured in becquerels per kilogram (Bq/kg), either in terms of wet or dry weight, based on the kind of specimen. The reported uncertainty is the mixed uncertainty, determined utilizing the error propagation law at a 95% confidence level. It depends on the relative standard uncertainties of various factors, including the specimen mass, net peak area, total energy peak efficiency, the interest radionuclide half-life, and emission probability. Figure 1 illustrates Canberra's NaI (TI) detector, and the setup utilized in the present investigation.

Figure 1. NaI (TI) detector and the data acquisition system (State Company for Foodstuffs Trading laboratory

Specific Activity Amount

The specific activity content measures a substance's radiation efficiency, expressed in Bq/Kg. It represents the anomalies number happening every second in a radionuclide. This connection is defined by Equation 1. (Yang, 2024; Krieger, 1981)

$$S.A=C= A / M \quad (1)$$

Whereas:

A is the activity which is identified directly from the system's screen in Bq,

M is the specimen mass (kg).

The gamma spectroscopy was performed using a Sodium iodide (NaI (Tl)) detector measuring 3" x 3". The detector was calibrated for 3600 seconds and was fully shielded with a lead shield. This setup was utilized to measure the amounts of Naturally Occurring Radioactive Materials (NORM) (Yang, 2024). The weight of every specimen is measured in kilograms. Due to the NaI (Tl) detector's low energy resolution efficiency, the radionuclide daughter was selected to measure the radioactivity amount for two series: ^{238}U and ^{232}Th. Specifically, nuclide ^{214}Bi has been selected to indicate the radioactivity for the ^{238}U series, while the radioactivity of the ^{232}Th series was identified using the radioactive isotope ^{228}Ac. Regarding ^{40}K, it may be immediately detected by the detector (Yang, 2024; Krieger, 1981; Mehra & Bala, 2014).

Radium Equivalent Activity (Ra_{eq})

Radiation hazards due to the specified radionuclides of ^{226}Ra, ^{232}Th and ^{40}K were measured by various indices. The radium equivalent activity is the most broadly utilized radiation index hazard (Ra_{eq}) (Zakaria et al., 2013). The radium equivalent activity is the weighted sum of activities of the above three radionuclides depending on the guesstimate that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of the ^{232}Th and 4810 Bq/kg of ^{40}K generate similar gamma-ray rates dosage, Ra_{eq} activity has been gained by (Nations et al., 2002):

$$Ra_{eq} = CU + 1.43\ CTh + 0.077\ CK \quad (2)$$

Whereas:

CK, CTh and CU are the activity amounts of ^{40}K, ^{232}Th and ^{238}U, respectively, in Bq/kg (Nations et al., 2002).

Dosage in Air, Annual Effective Dosage, and the Relating Internal and Exterior Indices

The total air absorbed rate dosage (D) since the average specific activity amounts of ^{238}U, ^{232}Th and ^{40}K (Bq/kg) has been determined by the following formula (Örgün et al., 2007).

$$D\ (nGy/h) = 0.427CU + 0.662CTh + 0.043CK \quad (3)$$

The yearly dosage equivalent (AEDE) received outdoors by an individual is determined by multiplying the absorbed dosage rate by the dosage conversion factor of 0.7Sv/Gy. The occupancy factor for outdoor exposure is 0.2 (5/24), while the indoor utilization factor is 0.8 (19/24). The determination of AEDE is based on the following equations (Oudah & Al-Mashhadani, 2015):

$$AEDE\ (Outdoor)(mSv/y) = Absorbed\ dosage(nGy/h) \times 8760h/y \times 0.7Sv/Gy \times 0.2 \times 10^{-6} \quad (4)$$

$$AEDE\ (Indoor)\ (mSv/y) = Absorbed\ dosage\ (nGy/h) \times 8760\ h/y \times 0.7Sv/Gy \times 0.8 \times 10^{-6} \quad (5)$$

The yearly effective dosage rate falls within the permissible dosage equivalent limit, ensuring safety and compliance with regulations. The formulas utilized for calculating the exterior index of hazards (H_{ex}) and the internal index (H_{in}) are as follows (Papastefanou et al., 2005):

$$H_{ex} = [CU/370(Bq/kg)] + [CTh/259(Bq/kg)] + [CK/4810(Bq/kg)] \quad (6)$$

$$H_{in} = [CU/180(Bq/kg)] + [CTh/259(Bq/kg)] + [CK/4810(Bq/kg)] \quad (7)$$

Another index of radiation hazard, known as representative index level (Iγr), was identified as the following (Pereira & Kelecom):

$$I\gamma r = CU/150 + CTh/100 + CK/1500 \quad (8)$$

The gamma index establishes a correlation between the yearly dosage rate resulting from the surplus of exterior gamma radiation induced by surface materials. This method is used as a screening method to detect materials that may pose a health risk once they are utilized in a building.

Results and Discussion

Some imported legumes and grains from specimens of different origins were analyzed to complete the monitoring program. The findings of the activity amount of ^{40}K, ^{137}Cs, ^{214}Bi (^{238}U series) and ^{228}Ac (^{232}Th series) are illustrated in Table 2. The activity amount magnitude (Bq/kg) in White beans specimens have been detected (188.45-231.64), (B.D.L (below detected level) -0.177), (B.D.L -22.59) and (B.D.L -0.15), respectively. In rice specimens, (10.43-16.59), (B.D.L -0.18), (9.54-19.792) and (B.D.L-0.95), respectively, were detected. In broken chickpeas, specimens have been detected (146.38-183.76), (B.D.L), (B.D.L -1.1) and (B.D.L -0.5), respectively. In lentils, specimens were detected (122.48), (0.22), (4.98), and (0.54), respectively. Finally, in flour specimens (26.59), (0.175), (9.48), and (5.54), all previous magnitudes in Bq/Kg units were detected. The radioactivity amounts gained in this investigation are below the world average magnitudes (Radhi et al., 2022; Sam & Abbas, 2001). The findings gained for the radium equivalent activity ranged in white beans, rice, broken chickpeas, lentils, and flour (14.724-40.426), (10.343-20.895),(11.986-26.723),(15.183) and (17.601) in Bq/Kg, respectively, (as illustrated Figure 2), which is well below the threshold magnitude of 370 Bq/kg (Radhi et al., 2022). In addition, in Table (3), the present magnitudes of indoor annual effective dosage equivalent in the above specimens were (0.0402-0.0961), (0.0222-0.0444), (0.0325-0.0651), (0.0380) and (0.0434) mSv/y, respectively. The outdoor annual effective dosage equivalent was (0.01006-0.02404), (0.00554-0.01111), (0.00812-0.01627), (0.00950) and (0.01086) mSv/y, respectively, as illustrated Figure 3 and 4. All magnitudes of indoor and outdoor annual effective dosage equivalent lower

than the world average magnitudes (0.45) mSv/y for indoors and 0.07 mSv/y for outdoors) (Scheibel et al., 2006). The magnitudes of exterior indices of hazard in specimens have been detected (0.0397-0.1092), (0.0279-0.0564), (0.0323-0.0722), (0.0410) and (0.0525), respectively. For internal indices of hazard, it was (0.0397-0.1736), (0.0551-0.1129), (0.0323-0.1081), (0.0552) and (0.0796), respectively. For another index of radiation hazard known as representative level index ($I_{\gamma r}$) has been detected (0.1271-0.3050), (0.0705-0.1415), (0.1026-0.2063), (0.1202) and (0.1363), respectively, as illustrated Figure 5, 6 and 7. All magnitudes of indices of hazard are less than the world permissible unity magnitude (Scheibel et al., 2006).

The magnitude of total air absorbed dosage rate in white beans, rice, broken chickpeas, lentils, and flour have been detected (8.202-19.606), (4.522-9.067), (6.625-13.270), (7.750) and (8.858) (nGy/h), respectively, as illustrated Figure 8. All magnitudes have been detected to be lower than the limit (1.0 mSv/y) mentioned by the ICRP.

Table 2. The radionuclides amount and radium equivalent activity

Specimen No	^{137}Cs Bq/kg	^{214}Bi Bq/kg	^{228}Ac Bq/kg	^{40}K Bq/kg	$^{226}Ra_{eq}$ Bq/kg
01 W	0.177	22.59	B.D.L	231.64	40.426
02 R	0.163	19.792	B.D.L	14.33	20.895
03 W	B.D.L	B.D.L	B.D.L	193.82	14.924
04 B	B.D.L	12.574	B.D.L	183.76	26.723
05 W	0.1	19.82	B.D.L	210.22	36.007
06 W	B.D.L	B.D.L	0.15	188.45	14.724
07 B	0.18	5.82	0.04	153.21	17.674
08 R	B.D.L	14.2	B.D.L	16.59	14.322
09 B	B.D.L	B.D.L	0.5	146.38	11.986
10 L	0.22	4.98	0.54	122.48	15.183
11 R	0.18	9.54	B.D.L	10.43	10.343
12 R	0.166	10.49	0.95	16.33	13.106
13 F	0.175	9.48	5.54	26.59	17.601
Allowed max. magnitude(Alharbi & El-Taher, 2013; Tawalbeh et al., 2011)	1000	30	32	400	370

Table 3. Dosages and indices of hazard

Specimen No	AEDE mSv/y		Hex	Hin	$I_{\gamma r}$	D nGy/h
	Outdoor	Indoor				
01 W	0.02404	0.0961	0.1092	0.1736	0.3050	19.606
02 R	0.01111	0.0444	0.0564	0.1129	0.1415	9.067
03 W	0.01022	0.0408	0.0403	0.0403	0.1292	8.334
04 B	0.01627	0.0651	0.0722	0.1081	0.2063	13.270
05 W	0.02146	0.0858	0.0973	0.1538	0.2723	17.502
06 W	0.01006	0.0402	0.0397	0.0397	0.1271	8.202
07 B	0.01116	0.0446	0.0477	0.0643	0.1413	9.099
08 R	0.00831	0.0332	0.0418	0.0823	0.1057	6.776
09 B	0.00812	0.0325	0.0323	0.0323	0.1026	6.625
10 L	0.00950	0.0380	0.0410	0.0552	0.1202	7.750
11 R	0.00554	0.0222	0.0279	0.0551	0.0705	4.522
12 R	0.00712	0.0285	0.0354	0.0653	0.0903	5.810
13 F	0.01086	0.0434	0.0525	0.0796	0.1363	8.858

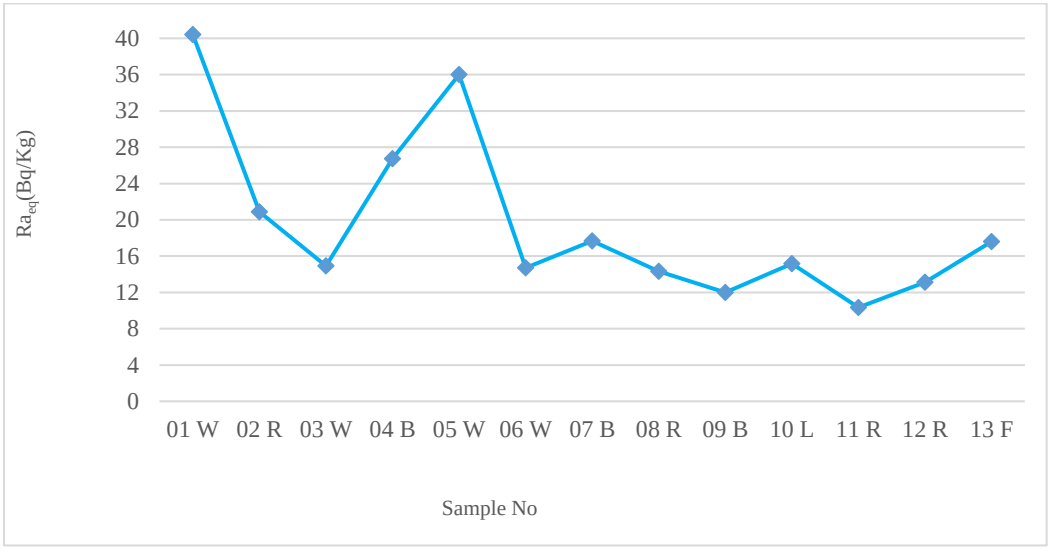

Figure 2. The relationship between Ra_{eq} and specimens no

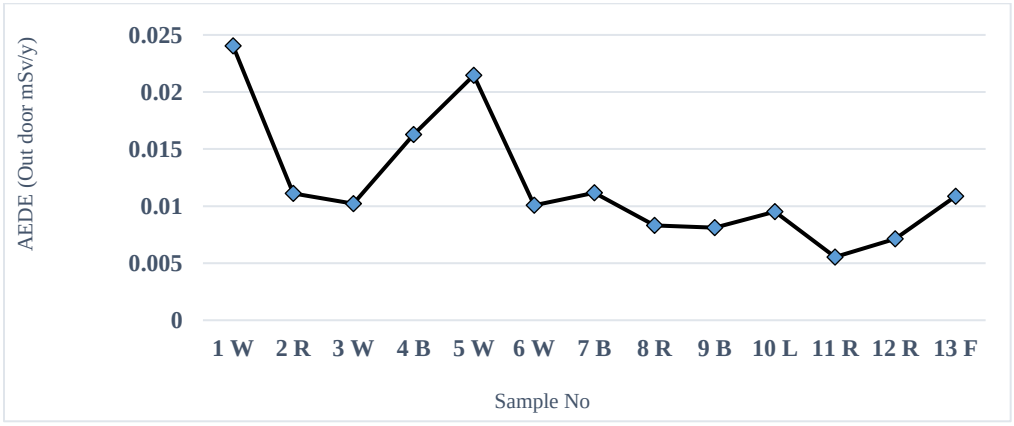

Figure 3. The relationship between AEDE outdoor and specimen no

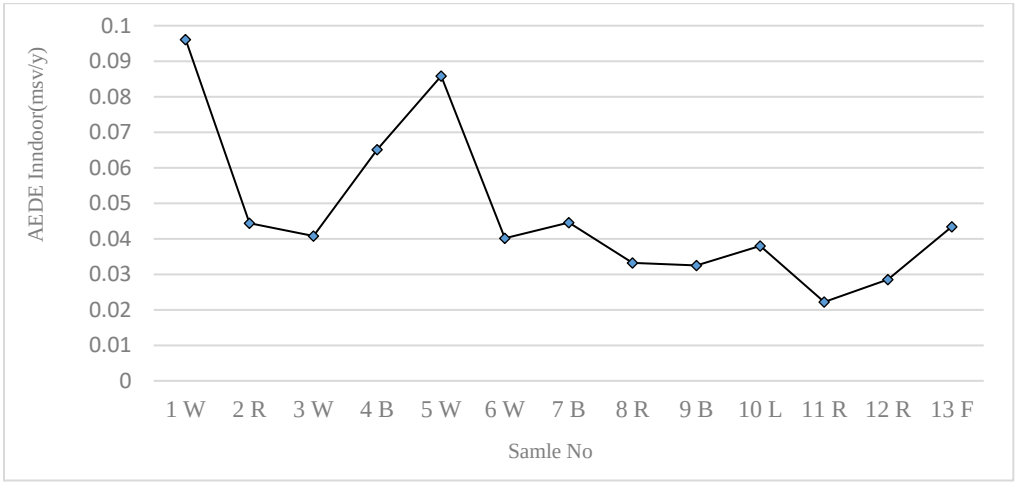

Figure 4. The relationship between AEDE indoor and specimen no

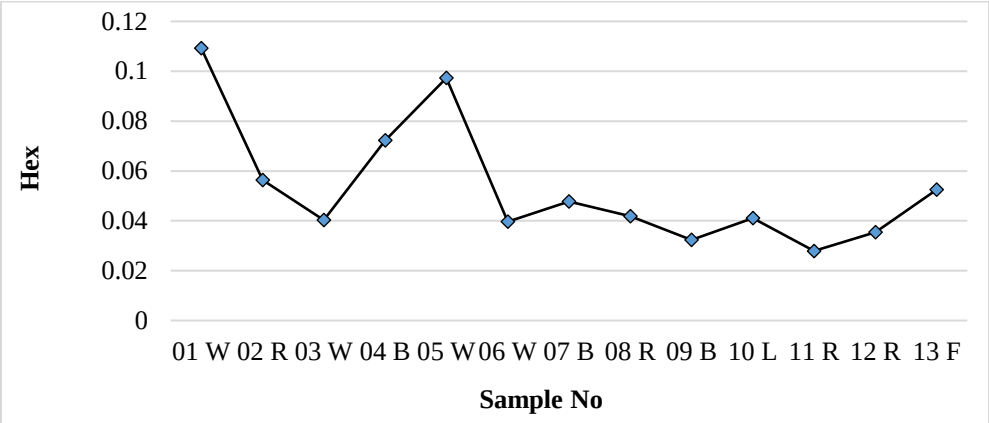

Figure 5. The relationship between Hex and specimens no

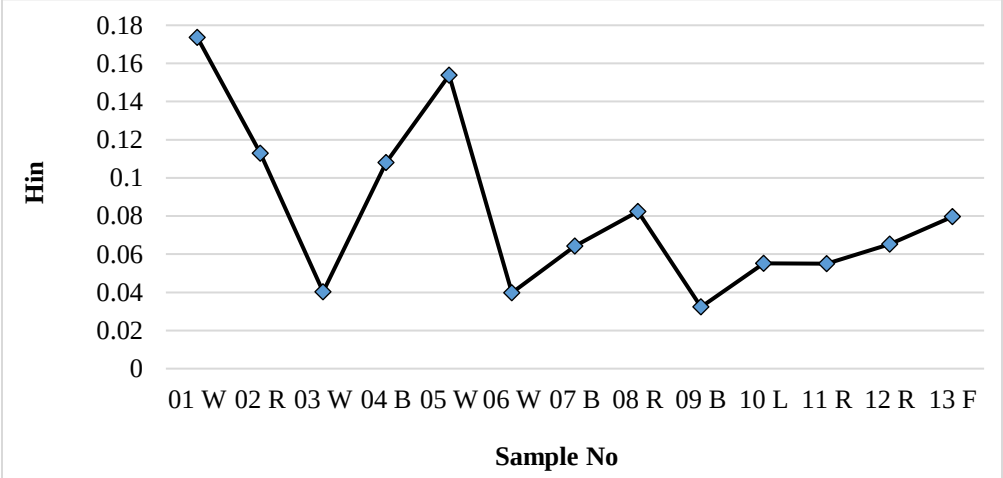

Figure 6. The relationship between Hin and specimens no

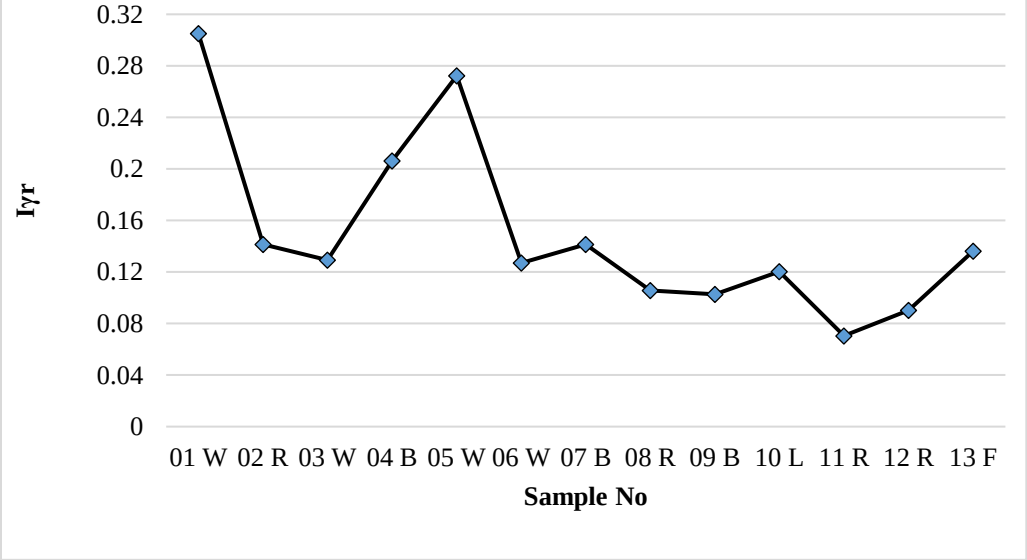

Figure 7. The relationship between Iγr and specimen no

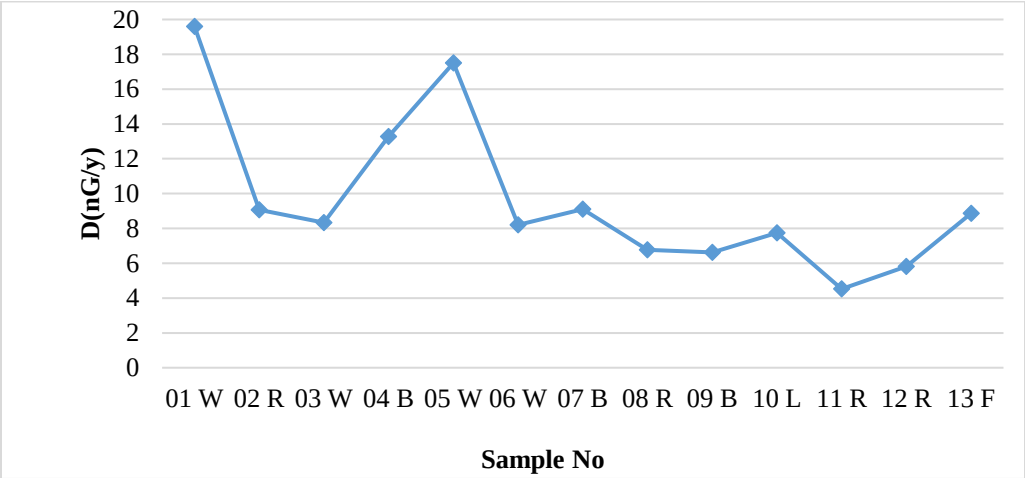

Figure 8. The relationship between D and specimen no

Conclusion

As mentioned above, the natural and anthropogenic radionuclides ^{40}K, ^{137}Cs, ^{214}Bi, and ^{228}Ac can be recognized in the spectra of legumes and grains. ^{226}Ra and ^{232}Th activity were calculated from ^{214}Bi and ^{228}Ac data, correspondingly presuming the worldly equilibrium among these testers.

The magnitudes of activity intended for ^{40}K, ^{137}Cs, ^{214}Bi and ^{228}Ac in legumes and grains testers are lower than the allowed world mean. The activity magnitudes for ^{137}Cs have been lower. Vice versa, the ^{40}K activity has been the uppermost. When potassium is consistently circulated in the body, it keeps being taken in foodstuffs, and its amount is based on homeostatic control. It has been at lower risk for human health than ^{137}Cs.

The magnitudes for the mean magnitude of annually total air absorbed rate dosage in these specimens were to be under a limit of (1.0 mSv/y) based on ICRP. The magnitudes for the (Ra_{eq}) have been within the international average allowable extreme magnitude of 370 Bq/kg. This investigation in this paper can be advantageous as a starting point for data on the amount of radionuclide and radium equivalent activity.

In general, the applied procedure has given worthy consequences than other reported data in the literature. In addition, the basket foodstuffs (legumes and grain) provided by the government to the Iraqi people here investigated have been safe for human intake due to radioactivity levels have been less than the permitted max levels.

Author Contributions

All Authors contributed equally.

Conflict of Interest

The authors declared that no conflict of interest.

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