Measurement of natural radioactivity concentrations in walnut collected from different markets in Sulaimanya city-Kurdistan region-Iraq

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Abstract: Walnut is one of the most important and necessary foodstuffs in our daily lives and there are many species in Iraqi Kurdistan from different sources. Therefore, the significant point in this works is gamma rays measuring to estimate various radiation hazards via walnut consumption for different markets. The samples have been collected from different markets and locations in Sulaimanya governorate in Kurdistan region-Iraq.

The average specific activities for nuclides found with high probability for the walnut by using a vertical coaxial cylindrical High Purity Germanium Radiation Detector (HPGe). The specific activity for the shell of the walnut were for the three most important radionuclides 232Th, 226Ra and 40K were 14.842 ± 9.450, 7.226 ± 1.805, 107.2 ± 2.563 (Bq/kg) respectively, and for walnut pulp 232Th, 226Ra and 40K were 16.053 ± 8.861, 5.105 ± 1.540, 143.6 ± 2.884 (Bq/kg) respectively. The total average activities were within the average ranges of the accepted values in the world (45, 33 and 420 Bq/kg for 232Th, 226Ra and 40K respectively).

1 – Introduction

Most fruits, vegetables, grains and foods are exposed to various radiations, both from terrestrial, atmosphere and from cosmogenic it is followed by a series of decay [1-3]. We swallow daily and eat many of the things that we do not know the content of radioactive materials, one of these is walnuts, which is a natural food for every family in Sulaimanya.

Neither one of the research institutions nor any of the researchers have tested the presence, types, quantity, and permissible doses of radioactive materials in the walnut in Sulaimanya. The concentrations of radioactive substances increase in soil and plants in which they are higher in the cultivated land that contains high concentrations of radioactive materials [4]. The most radioactive nuclides sources of external radiation on earth in food and water are 232Th, 40K, 226Ra and fall out radioisotope 137Cs and the results indicate that the presence of radionuclides in drinks is higher than that in food [5].

Knowing the concentrations and permissible dosages of radioactive materials is important to know the harmful limits for humans [6] because they pass through eating it to our bodies and may be important sources of internal dose for people living in the region which make particular hazards [7]. Some geographical and terrestrial factors affect the ranges of concentrations of radioactive materials and the activity level are strongly depending on the origin of the ores. [8, 9].

Several factors contribute to the amount of radioactive nuclides taken from plants such as plant age or the soil kind [9, 10]. Radioactive nuclides remain in the layer of cultivated soil in so called
active zone of plant roots for many years [11]. The radioactivity levels in food grown in such soils may also increase, through various uptake mechanisms from soil to plant [12].

Walnut for consumption among Sulaimanya governorate in Kurdistan region of Iraq, mainly comes from imported walnut and considered an essential part of food, it imports from Hauraman/ Sulaimanya, Chilly, USA, Romania and Ukraine and other sources. The primary purpose of this study is, therefore, to determine the activity concentration levels of radionuclides in walnut and compare the results with natural radionuclide references.

A total of five kilograms of Walnut from different markets and several sources was collected in Sulaimanya city in Kurdistan region-Iraq, for analysis and to quantify the presence of natural radionuclides in it and to determine the activity concentrations and their corresponding hazard.

2 – Materials and methods

The walnut samples were collected from five different locations in Sulaimanya. Special type beakers (0.6 kg in a 1 liter Marinelli beaker) were used for processing and measuring the samples. The net weights of the samples were found from the difference between the weights of a sample-filled and empty beakers.

Finally, the beakers filled with samples and closed with caps and kept for one month for achieving the secular equilibrium.

The walnut core samples were isolated from the crusts, and then the shell samples were grinded well to become powder until they become homogeneous and placed in equal Marinelli beakers for 30 days to reach the secular radioactive equilibrium condition. A measurement of radioactivity and radionuclides in the samples was carried out by gamma spectrometry analyzer [1]. Using a vertical coaxial cylindrical coaxial High Purity Germanium Radiation Detector (HPGe) manufactured by Princeton Gamma Tech (PGT) based on a coaxial p-type with large crystal diameter 70.6 mm and length 70.7 mm. The detector specified with accurate energy resolution, full width at half maximum (FWHM), 1.1 keV for the energy 122 keV $^{57}$Co and 1.97 keV for the energy 1332 keV $^{60}$Co gamma-ray line and relative efficiency higher than 70% at 122 keV ($^{57}$Co) as shown in figure (1). A high voltage power supply generates a voltage ranged (0–4000 V) to feed the detector through a preamplifier. Subsequently, the detector connected to a chain of electronic units for completing signal processing such; Main amplifier, Analogue to digital converter (ADC), Multichannel analyzer (MCA) and Commercial software Quantum Gold from Princeton Gamma-Tech (PGT) was used for data analysis that interfaced on PC. Gamma spectrometry was shielded by a thick shield (5 cm) of lead encasing the detector (10cm inner diameter, and 50cm of height).

![Figure 1](image.png)

**Figure 1.** Relative efficiency per energy peaks of the standard sources
The environmental background radiation achieved by the blank sample was subtracted from each spectrum of the samples [2]. The system is located inside cylindrical lead shield Kolga Model A340 about 10.1 cm high-performance copper/tin lined lead shield for Germanium radiation detectors to reduce the background scattering for the measured spectrum. Reducing the leakage current as a result of thermal noise of the detector has been done by cooling it with liquid nitrogen. This shield is eliminating the contribution of laboratory radiation background to the sample activity.

The analysis process was fixed for the duration of 6 hours to outcome gamma spectrum information that agrees with previous studies [13]. Determining the counting time to eliminate the error of counting statistics to less than 5% in the measured net peak area of the major gamma photo-peaks for 60Co. The samples were then placed on the top of the detector and were counted for 6 hours in an attempt to eliminate a counting error. Samples are 0.6 kg in a 1 liter Marinelli beaker which was designed to specially fit over a coaxial detector end cap so that the sample essentially surrounds the detector which is useful for the analysis of low-activity samples [3, 4]. The activity measurements of radionuclides results of the samples were used to calculate, activity concentrations of the radionuclides, the investigated level gamma indexes, radium equivalent activity, the absorbed gamma radiation dose and the effective dose exposure.

The photopeak at 1461 KeV was used for the measurement of 40K, whereas 186.1 KeV and 63.8 KeV, were used for the measurement of 226Ra and 232Th, respectively.

2.1 – Efficiency and Energy Calibration

The energy and efficiency calibrations were carried out before analyzing process of the samples as previously reported [14]. The calibrations of gamma energy and efficiency calibration of the system were found by some standard sources with known energies, such as 60Co to derive the calibration curve for energy and the efficiency of the gamma-ray spectrometry and energy that is in agreement with the study of [15]. HPGe- system distinguished by using the multi peak of the gamma ray line for the determination of the 226Ra and 232Th activities, and to obtain more accurate results for determining other extra radionuclide of the samples.

2.2 – Activity concentrations and Gamma Dose Rate

The specific activity or the activity concentration $A$ of the radionuclides defined as the activity per unit mass of the sample (Bq/kg) and were determined using the following equation [16]:

$$A = \frac{C_n}{T_{\gamma} M \epsilon(E_{\gamma}) t_c}$$  \hspace{1cm} (1)

where $C_n$ is the net count area per second (Total counts minus background counts), $T_{\gamma}$ Is the absolute transition probability of the specific gamma, $M$ is the mass of the sample (kg), $\epsilon(E_{\gamma})$ Is the HPGe detector efficiency in energy $E_{\gamma}$ of the specific gamma energy and $t_c$ is the counting time.

The investigated level gamma indexes ($I_{\gamma}$), used for the estimation of gamma radiation, is derived from the specific activity ($A_c$) in Bq/kg. For 226Ra, 232Th, and 40K, it was calculated using the following equation as reported in European Commission [17]:

$$I_{\gamma} = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_{K}}{3000}$$  \hspace{1cm} (2)

where $I_{\gamma}$ Is: activity concentration index, it is unitless and has a unitary value and its value must be equal or less than one. $A_{Ra}$, $A_{Th}$ and $A_{K}$ are the radium, thorium and potassium activity concentrations (Bq/kg).

A common radiological index that represents the activity concentrations and existing of natural radionuclide (226Ra, 232Th and 40K) in the terrestrial is called radium equivalent activity (Ra$_{eq}$) in Bq/kg to ensure the uniformity in the distribution of these natural radionuclides and this allows the comparison of specific activity of these materials that have different quantity of the three types of radionuclides and is given by [18-19]:

$$Ra_{eq} = A_{Ra} + 1.43 A_{Th} + 0.077A_{K}$$  \hspace{1cm} (3)
Where, $A_{Ra}$, $A_{Th}$ and $A_{K}$ are the activity concentrations of the radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K in Bq/kg respectively. The ratios of natural radionuclides are: 10 Bq/kg for $^{226}$Ra, 7 Bq/kg for $^{232}$Th and 130 Bq/kg for $^{40}$K. These ratios are participating in formulating $R_{eq}$, in which its maximum limit is 370 Bq/kg [20].

The absorbed gamma radiation dose from the air is calculated from the substrate at a height of 1 meter and is measured in nGy/h [14]. The sub-activities must be multiplied by a conversion factor in which determined for the average concentration of radionuclides from each radioactive series.

$$D = (R_{Ra}.A_{Ra}) + (R_{Th}.A_{Th}) + (R_{K}.A_{K})$$

Where: $D$ is the absorbed dose rate of gamma radiation from the air(nGy/h), $R_{Ra}$, $R_{Th}$ and $R_{K}$ are the conversion factor for $^{226}$Ra, $^{232}$Th, $^{40}$K respectively, in which equals to 0.4551, 0.5835, 0.0429 (nGy hr$^{-1}$/Bq kg$^{-1}$) respectively, as given [20], therefore:

$$D(nGy/h) = 0.4551 A_{Ra} + 0.5835 A_{Th} + 0.0429 A_{K} \quad (4)$$

$A_{Ra}$, $A_{Th}$, and $A_{K}$, are the activity concentrations (Bq/kg) for $^{226}$Ra, $^{232}$Th and $^{40}$K respectively [21].

The effective dose exposure to the human in one year due to the soil radioactivity can be estimated by using a conversion factor of (0.7 Sv/Gy) recommended by the UNSCEAR report [20]. The annual effective dose rate ($H_{ann}$) in unit (mSv/y) is the best radiation risk indicator in which calculated from [20, 22-23]:

$$H_{ann}(mSv/y) = H(nGy/h) \times (365 \times 24)h \times 0.7 \text{ Sv/Gy} \times 10^{-6} = 0.006132 \times H(nGy/h) \quad (5)$$

The conversion coefficient for the absorbed dose in air to the received effective dose is 0.7Sv/Gy. The outdoor (or indoor) $H_{ann}$ calculate by multiplying the above equation by 0.2 for outdoor and 0.8 for indoor, where the most people spent their times ~20% in outdoor.

Another important factor for internal exposure to the radio nuclides, is the internal hazard index $H_{in}$, in which given by the following equation [14,22]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \quad (6)$$

$H_{in}$ must be less than unity (100%), for providing safety level and for protecting people. External hazard index $H_{ex}$ equation is the same as $H_{in}$, but the only difference between the two equations of $H_{in}$ and $H_{ex}$, is the number 185 replaced by 370.

**Results and discussion:**

In this research, the samples of walnut were divided into two parts (core and shell), and each one of them were checked and analyzed. Table (1) shows the specific activity of natural ($^{226}$Ra, $^{232}$Th, $^{40}$K) radionuclides in walnut, in which the mean values from references [24-26] were tabulated also within it. The activity concentration for $^{226}$Ra, $^{232}$Th and $^{40}$K varied from (2.725 ± 1.853) to (7.226 ± 1.805) Bq/kg, from (0) to (16.053 ± 8.861) Bq/kg, and from (31.789 ± 1.329) to (143.6 ± 2.884) Bq/kg respectively as shown in figures (2–4). The average value of specific activity of natural $^{226}$Ra, $^{232}$Th, $^{40}$K and $^{137}$Cs radionuclides in walnut were, 4.6538, 5.1491 and 98.5789 Bq/kg, respectively. They were more less than the permitted mean values (50, 15 and 382 Bq/kg) of mentioned natural radionuclides [24-26], that is mean that there is no any hazard with these samples of walnuts.

The average value of specific activity of the (core) in walnut for natural $^{226}$Ra, $^{232}$Th and $^{40}$K radionuclides were, 3.318, 8.0265 and 129.3 Bq/kg but for them (shell) were respectively 5.9894, 7.421 and 67.8578 Bq/kg, that was indicated that the average value of the walnut core for $^{40}$K, was higher than that of the shell. The maximum value of specific activity of $^{226}$Ra, was found to be (5.105 ± 1.540) Bq/kg for sample 3 which is the core of Ukraine walnut type, but its maximum value of the shell was (7.226 ± 1.805) Bq/kg for the (sample 7-USA type), therefore the specific activity of the
shell is greater than that of the core but still less than the average permitted value (50 Bq/kg). The maximum recorded specific activity value for $^{232}$Th, was $(16.053 \pm 8.861)$ Bq/kg (sample 3-Ukraine type-core) and $(14.842 \pm 9.45)$ Bq/kg for (sample 7, USA type-shell) but not exceed the permitted value 15 Bq/kg. The maximum specific value for the $^{40}$K radionuclide was $(135.7 \pm 2.835)$ Bq/kg for the core of (sample 4 Romania type) and $(107.2 \pm 2.563)$ Bq/kg for the shell of (sample 6-Chili type) in which it is less than the average value 382 Bq/kg [24].

From table (2) and figure (5) show that the radium equivalent activity ($Ra_{eq}$) range is between 7.887 and 37.631 Bq/kg with a mean value of 16.6606 Bq/kg, which is below the average value, 18.98 Bq/kg [26]. On the other side of the same figure (5), the absorbed dose rate $D$ of gamma radiation for all samples, has maximum value 17.022, which is more less than the mean standard value 59.293 nGy/h, therefore there is no any hazard.

The recorded values of the investigated level gamma indexes ($I_{\gamma}$) and internal hazard index $H_{\text{in}}$, were tabulated in table (2) as shown in figure (6). It has been shown that the maximum value of $I_{\gamma}$ was 0.5275 (52.75%) for sample 4 (RO4-N) which is for the walnut core and it is greater than the exceed permitted value which is 0.164 (16.4%). Internal hazard index $H_{\text{in}}$ has been calculated also as shown in table (2) with figure (5). The maximum value of $H_{\text{in}}$ is 0.1154 (11.54%) for sample number 3 (UK3-N) which is in the core zone of the walnut, but still much less than the permitted value 0.94 (94%). As a result for all, this research denotes that these samples of walnuts are safe for eating that, for Romania (core) sample is greater than the average value. Besides that the $Ra_{eq}$ (Bq/kg) for both USA (core) and USA (shell) are; 37.631 and 33.933 (Bq/kg) respectively, which are also greater than the average value recorded. Determination of the minimum detectable activity (MDA) of a radionuclide $^{137}$Cs being assessed through gamma ray spectrometry and also tabulated in table (1).

**Table 1.** Specific activity of the radionuclides $^{226}$Ra, $^{232}$Th and $^{40}$K with the Mean values.

| Sample no. | Sample ID | Sample Name    | Specific activity( Bq/kg ) | $^{226}$Ra | $^{232}$Th | $^{40}$K | MDA (Bq/kg) |
|------------|-----------|----------------|-----------------------------|------------|------------|----------|-------------|
| 1          | SH1-N     | Chili (core)   | 3.035 ± 2.032               | 0.00       | 143.6 ± 2.884 | 1.75E-02 |
| 2          | AM2-N     | USA (core)     | 2.725 ± 1.853               | 0.000 ± 0.000 | 131.0 ± 3.011 | 2.29E-02 |
| 3          | UK3-N     | Ukraine (core) | 5.105 ± 1.540               | 16.053 ± 8.861 | 124.3 ± 2.801 | 2.01E-02 |
| 4          | RO4-N     | Romania (core) | 2.811 ± 0.926               | 0.00       | 135.7 ± 2.835 | 2.53E-02 |
| 5          | KH5-N     | Sharbazer (core) | 2.914 ± 1.211            | 0.00       | 111.9 ± 2.703 | 2.12E-02 |
| 6          | SH1-T     | Chili (shell)  | 4.415 ± 1.586               | 0.000 ± 0.000 | 107.2 ± 2.563 | 2.25E-02 |
| 7          | AM2-T     | USA (shell)    | 7.226 ± 1.805               | 14.842 ± 9.450 | 71.21 ± 2.395 | 1.97E-02 |
| 8          | UK3-T     | Ukraine (shell) | 5.924 ± 1.393               | 0.000 ± 0.000 | 62.21 ± 2.107 | 0.00       |
| 9          | RO4-T     | Romania (shell) | 6.933 ± 1.120               | 0.00       | 66.88 ± 1.966 | 1.76E-02 |
| 10         | KH5-T     | Sharbazer (shell) | 5.449 ± 1.437              | 0.000 ± 0.000 | 31.789 ± 1.329 | 2.15E-02 |
|            | Median    | 50             | 15                          | Maximum    | 382 [24]    | Median < 0.4 [25] |
Table 2. Gamma indexes ($I_{\gamma}$), Radium equivalent ($Raeq$), absorbed dose rate (D) and internal hazard index ($H_{in}$) for all samples with the mean values.

| Sample no. | Sample ID | Sample Name        | $I_{\gamma}$ | $Raeq$(Bq/kg) | D(nGy/h) | $H_{in}$ |
|------------|-----------|--------------------|--------------|---------------|----------|----------|
| 1          | SH1-N     | Chili (core)       | 0.05766      | 14.092        | 7.541    | 0.0462   |
| 2          | AM2-N     | USA(core)          | 0.5275       | 12.812        | 6.860    | 0.0419   |
| 3          | UK3-N     | Ukraine(core)      | 0.1387       | 37.631        | 17.022   | 0.1154   |
| 4          | RO4-N     | Romania(core)      | 0.5460       | 13.259        | 7.100    | 0.0434   |
| 5          | KH5-N     | Ukraine(Shell)     | 0.0470       | 11.530        | 6.126    | 0.0390   |
| 6          | SH1-T     | Chili (shell)      | 0.0504       | 12.669        | 6.608    | 0.0461   |
| 7          | AM2-T     | USA(Shell)         | 0.1221       | 33.933        | 15.003   | 0.1111   |
| 8          | UK3-T     | Ukraine(Shell)     | 0.0404       | 10.714        | 5.364    | 0.0449   |
| 9          | RO4-T     | Romania(Shell)     | 0.0453       | 12.079        | 6.024    | 0.0513   |
| 10         | KH5-T     | Ukraine(Shell)     | 0.0287       | 7.887         | 3.887    | 0.0360   |
|            | **Mean value [27]** |                | **0.164** | **18.98** | **59.293** | **0.94** |

Figure 2. Specific activity of radionuclides $^{226}$Ra for the samples.

Figure 3. Specific activity of radionuclides $^{232}$Th for the samples.
Figure 4. Specific activity of radionuclides $^{40}\text{K}$ for the samples.

Figure 5. Radium equivalent activity (Ra$_{eq}$) ranges and the absorbed dose rate of gamma radiation for the samples.

Figure 6. Activity concentration index $I_{\gamma}$ and internal hazard index $H_{in}$ for the samples.
Conclusion

In this study, activity concentrations for walnut has been studied and it was found that all of its values for the three radionuclides $^{226}$Ra, $^{232}$Th, $^{40}$K and $^{137}$Cs were under the average values. Gamma indexes ($I_{\gamma}$), the average value of radium equivalent activity ($Ra_{eq}$), absorbed dose rate ($D$) and internal hazard index ($H_{in}$). For all samples were with a good agreement with the national values except, there were two values of gamma indexes ($I_{\gamma}$) was higher than the mean value for sample 2 (the core of the USA one) and sample 4 (the core of Romania type). Besides that the $Ra_{eq}$ (Bq/kg) for both USA (core) and USA (shell) are, greater than the average value recorded.

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