Assessment the Natural Radioactivity of Radionuclides \((226^{\text{Ra}}, 232^{\text{Th}}, 40^{\text{K}}, \text{and } 137^{\text{Cs}})\) in Wheat Grain

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Abstract—This paper investigates the activity concentration of radionuclides \((226^{\text{Ra}}, 232^{\text{Th}}, 40^{\text{K}}, \text{and } 137^{\text{Cs}})\) in the wheat grain samples using a high-purity germanium detector. Thirty-six wheat grain samples were collected from different locations of Koya City, Iraqi Kurdistan region. Average activity concentrations of \(^{226}\text{Ra}, ^{232}\text{Th}, \text{and } 40^{\text{K}}\) in wheat grain are found to be 0.407 ± 0.097 Bq.kg\(^{-1}\), 0.36 ± 0.14 Bq.kg\(^{-1}\), and 109.25 ± 2.214 Bq.kg\(^{-1}\) for \(^{226}\text{Ra}, ^{232}\text{Th}, \text{and } 40^{\text{K}}, \text{respectively.}\) The measured activity concentrations for the radionuclides are compared with the reported data from other countries. In addition, the fallout radionuclide of 137Cs has no detection of in the wheat grain samples. The radium equivalent activity Ra\(_{eq}\), internal and external hazard indices H\(_{in}\) and H\(_{ex}\), and annual gonadal dose equivalent are calculated for the measured samples. The total ingestion dose is 113.19 μSv.y\(^{-1}\), which is below the world average value of 290 μSv.y\(^{-1}\).

Index Terms—Natural Radioactivity, Wheat Grain, Radionuclides, \(^{226}\text{Ra}, ^{137}\text{Cs}\).

I. INTRODUCTION

Natural sources of ionizing radiation have continual property of emission of nuclear particles or Gamma-rays, therefore, the exposure to human beings by those sources of radiation is inescapable. The primordial radionuclides comprise the natural series such as \(^{238}\text{U}, ^{232}\text{Th}, \text{and non-series } 40^{\text{K}}\) which are ordinarily long lived and with a half-life more than one hundred million years (Al-Hamzawi, 2017a, UNSCEAR, 2000). The radionuclide radiation could be a serious problem to the living tissues, because can cause damage them just when the radiation energy is absorbed in that tissues, and food ingestion is the most common pathway to transfer radionuclides to people, therefore, the detection of radioactive materials is absolutely important in the process of people and environment protection (Harb, et al., 2014).

Uranium and its isotopes are considered most serious pollution due to its radiological and toxicological activity which is a threat to the human and the environment, the ingestion of food is considered the main pathways of uranium entrance into the human body (Zakariya, 2019). Human beings are exposed to both external and internal radiation. The internal exposure comes from the intake of terrestrial radionuclides through inhalation or ingestion pathway. The inhalation exposure is related to the existence of dust particles in the air which comprise the radionuclides from the decay series of \(^{238}\text{U}\) and \(^{232}\text{Th}\) and non-series \(^{40}\text{K}\) as well. Plants acquire the main source of natural background radiation (terrestrial radionuclides) through the roots and leaves whereas humans and animals acquire radionuclides through consumption of these plants, there are two different mechanisms for the transferring of radionuclides to plants, either through root uptake or directly through aerial deposition (Khan, et al., 2011).

The levels of radionuclides in plants vary typically from a few tens of Becquerel (Bq) to several hundred of Becquerel per kilogram (Wang, et al., 1997). The radionuclides that exist in the fertilizers are uranium and thorium decay series as well as potassium. Besides, the concentration of radionuclides in fertilizers differs from different countries and depending on the origin of the components. Measurement and assessment of natural radioactivity is necessary because of its immediate effect on the human beings safety. In the most countries of the world, the study of naturally occurring radiation and environmental radioactivity was carried out (UNSCEAR, 2000). During the past decades, the agricultural activities in Iraqi Kurdistan region widely grew up, especially wheat planting due to the application of different types of 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 (Brigden, et al., 2002). From many countries, to establish a baseline data to the natural radioactivity levels, measurement of natural radionuclides in environmental elements has been carried out (Zakariya, 2019).

Therefore, this research was carried out to investigate the levels of radioactivity due to the natural radionuclides of \(^{226}\text{Ra}, ^{232}\text{Th}, ^{40}\text{K}, \text{and } 137^{\text{Cs}}\) in wheat grain of Koya City,
Iraqi Kurdistan, and also to estimate the radiological hazard parameters of wheat grain samples.

II. RESEARCH METHODOLOGY

A. Study Area

This study was conducted at Koya (Koysinjaq) in Erbil governorate from the south part of Iraqi Kurdistan region, as shown in Fig. 1. Koya district is about 582 m high from sea level, and it’s geographical coordinates are 36.0751° N and 44.6199° E. Furthermore, Koya is a mountainous area, and it is surrounded by many villages which have affected the lifestyle of the people that living there. In general, it is considered as a good agricultural region. The intensive farming of wheat is distributed at the plain of Erbil, south of Koysinjaq district (Zakaria, et al., 2013, Hussein, 2015). The proper location and weather of this district (rainy winter and hot summer) are two helpful parameters in growing up the agricultural activities there. Especially, wheat planting has attracted a lot of attention of the farmers due to the facility in planting and it’s well growing in that region (Salih, et al., 2020a). So that, wheat production can be considered as a dominant agricultural activity in that district and the largest area of agricultural lands is devoted for wheat planting (86.4%, 171,750 donums). As well as, it is estimated that wheat covers the most portion of farmlands in the world (Servitzoglou, et al., 2018). Therefore, this study was done to estimate the concentration levels of natural radionuclides and the radiological hazards in wheat grains resulting from consumption of wheat flour in Koya district.

B. Radioactivity in Wheat Plant

One of the most important food crops in the world is wheat. Annually, the largest agricultural area is devoted to wheat plantation. Wheat is a stable daily food in many different forms. In Kurdistan region, an ample amount of wheat is consumed in the form of flat bread which is locally called Nan. It is known that the major fraction of the radioactivity is retained by root part of the wheat plant. Some fraction of the radioactivity is up taken by the grain part of the wheat from the soil (Chen, et al., 2005). A part of the radionuclides which present in the fertilized soils could be taken by the plants through root uptake. Then, they can be transferred to the human body by food ingestion. The radiation dose rate taken by the human body through the different organisms depends on several factors; the rate of food consumption, the soil characteristics which the particular crop has grown on it, the health and how old is the user (Tsukada, et al., 2002). Depending on their requirement, the plants may take up the nutritious ions then they are transferred to particular tissues according to the function of the element in plant metabolic process. The primordial radionuclides could also be transported along with nutrients and may have the same chemical behavior as the indispensable nutrient. The distribution of $^{238}$U and $^{232}$Th in various parts of the wheat plant there is a decreasing trend as; root> shoot> husk> grains, radionuclides have the lowest concentration in the wheat grains and about 50% of Ra is observed to pile up in the roots and nearly 22% in the shoots and husk. From the figure, it is also could be seen that the higher concentration of $^{40}$K is in the shoots and it follows a decreasing trend as shoot> root> husk> grain (Pulhani, et al., 2005).

C. Wheat Grain Sampling

A total of 36 samples of mature wheat grains obtained from the wheat plants grown were collected at harvesting time during, among the center of Koya district and it’s five subdistricts (Ashti sub-district, Taq taq sub-district, Segirdkan sub-district, Shorsh sub-district, and Siktan sub-district) within 36 villages where the local growers use a great area of land for the cultivation of wheat plant. The sample locations are shown in Fig. 2. To make a representative sample from each location, 6 points were selected across each wheat plantation field, and the area of each point was 2 m × 2 m (IAEA, 1989). The wheat grain samples were labeled and transferred into a polythene bag. Thereafter, the samples were transported into the laboratory of research at Koya University. After then, the samples were carefully cleaned from wheat roots, wheat leafs, and any kind of debris. Then, the samples were crushed using a powder grinder machine (Silver Crest, model No.: SL-8859) and passed through a 1 mm mesh to get homogenized samples. To remove moisture and for adequate drying, the samples were placed in an electrical oven at 100°C for 10 h (Alshahri, 2016). A very sensitive balance was used to measure the mass of the dried samples, each sample about 1 ± 0.02 Kg of dry weight. For measurements, the samples were packed into standard size containers (Marinelli beakers) and tightly sealed then stored for a month to reach secular equilibrium (Zakariya, 2015). Finally, after ensuring that the radioactive equilibrium of the decay products of $^{226}$Ra and $^{232}$Th series reached by storage for 30 days (Cevik, et al., 2007). The stored samples were transported into the counting room (Nuclear Physics Laboratory – Koya University) for measurement and analysis.

D. Efficiency Calibration

The efficiency of a detector is ratio of the number of pulses recorded by the detector to the number of gamma-ray photons emitted by the source. Efficiency is the most important characteristic of the detectors, so that, a precise efficiency calibration of a gamma-ray spectrometry system
is necessary for the analysis of radionuclides available in a sample (Mostajaboddavati, et al., 2006). The calibrations of efficiency calibration for the system were performed using standard sources from the International Energy Agency (IAEA) as a function of gamma-ray energies. The detector has a relative efficiency of 73.8% at 1.33 MeV for $^{60}$Co, and its resolution (FWHM) was 1.18 keV at 122 keV for $^{57}$Co, and at 1332 keV of $^{60}$Co was 1.97 keV, the radioactivity measurements were carried out for 36,000 s (Essiett, et al., 2015, Salih, et al., 2020). The efficiency calibration of the gamma-ray spectrometry study was performed using $^{226}$Ra (186.1, 295, 351.9, 609, 665, 1120, and 1764 keV), $^{60}$Co (1175.2 and 1332.5 keV), and $^{137}$Cs (661.7 keV). The relative efficiency curve of the detector was made of the different energy values covering the energy range from 186 keV to 1332.5 keV. The efficiency calibration curve of high-purity germanium detector is shown in Fig. 3.

E. Calculation of the Activity Concentration of Radionuclide and Hazard Indices

After storing the samples for a month and under the assumption that secular equilibrium was achieved between $^{226}$Ra and $^{232}$Th and their decay products, the activity concentration of $^{226}$Ra was calculated from the average concentrations of $^{214}$Pb and $^{214}$Bi decay products and that for $^{232}$Th was calculated from the average concentrations of $^{208}$Tl and $^{228}$Ac decay products in the sample that is agree with AL-harbiI and El-Taher, 2013.

Activity concentration of radionuclides

The activity concentration of the interested radionuclides $^{226}$Ra, $^{232}$Th, $^{40}$K, and $^{137}$Cs in a unit of Bq.kg$^{-1}$ has been calculated using the relation (Murtagda, et al., 2017, Salih, et al., 2020).

$$\text{Activity concentration} \left( \text{Bq kg}^{-1} \right) = \frac{\text{Net count}}{\varepsilon \times I_{\gamma} \times t \times m} \quad (1)$$

Where, $I_{\gamma}$ is the emission probability per decay of the specific peak, $\varepsilon$ is the absolute gamma peak efficiency for the detector at a particular photopeak, $t$ is the counting time in seconds, and $m$ is the mass of the sample in kilogram.

Hazard indices

The exposure to radiation arising from the primordial radionuclides of $^{226}$Ra, $^{232}$Th, and $^{40}$K in the wheat grains can be determined in terms of some parameters, as given below; Radium equivalent activity ($Ra_{eq}$)

The radium equivalent activity ($Ra_{eq}$), which is a single index, used to describe the gamma output from different mixtures of $^{226}$Ra, $^{232}$Th, and $^{40}$K in the material. It was calculated from this equation (Nisar, 2015; Al-Hamed, et al., 2017).

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_{K} \quad (2)$$

Where, $A_{Ra}$, $A_{Th}$, and $A_{K}$ are activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K, respectively.

Internal hazard indices ($H_{in}$)

Internal hazard index of the gamma-ray specific activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K is calculated using Equation (3) given by (Ismail et al, 2020, Mehra, et al., 2007).

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \leq 1 \quad (3)$$

External hazard indices ($H_{ex}$)

The external hazard index is a description that quantifies the exposure factor and is an estimation of the hazard of the
natural gamma radiation due to the terrestrial radionuclides of $^{226}$Ra, $^{232}$Th, and $^{40}$K. It can be calculated using Equation (4) (Taiwo, et al., 2014).

$$H_{EI} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \leq 1$$  \hspace{1cm} (4)

Ingestion dose ($E_{ING}$)

Annual ingestion dose: The annual ingestion dose ($E_{ING}$) for human was coming from consumption of grain, due to the ingestion of radionuclides. The concentration of naturally occurring radionuclides of $^{226}$Ra, $^{232}$Th, and $^{40}$K in foodstuffs and the consumption rate of food and water by human beings affect the annual ingestion dose rates (UNSCEAR, 2000). The annual ingestion dose for Koya inhabitants ($E_{ING}$) coming from the consumption of wheat was calculated using the following equation given by (Canbazoglu and Dogru, 2013, Salih, 2018).

$$IAED = \sum i(li \times Ai, r) \times FDC_r$$  \hspace{1cm} (5)

Where, $i$ represents a food category (grain, vegetable, fruits, etc.); $li$ and $Ai$, $r$ represent the annual consumption rate of plant crops per capita (kg/year) and the activity concentration of radioactive nuclide $r$ in food category $i$ (Bq/kg), respectively, and FDC$r$ is the dose conversion factor for the ingestion of radionuclide $r$ (Sv/Bq). The committed DCFs for the radioactive nuclides in adults were obtained from the ICRP 2012 reported as 0.28, 0.23, and 0.006 µSv/Bq for calculations of the effective dose due to $^{226}$Ra, $^{232}$Th, and $^{40}$K, respectively (Murtadha, et al., 2017).

Annual gonadal dose equivalent (AGDE)

Gamma radiation affects various organs of the human body depending on the type of the organs and the duration of exposure. The most sensitive organs interested by UNSCEAR, 2010, are the bone surface, bone marrow, lungs, thyroids, female’s breast, and the gonads. Therefore, the AGDE due to the natural radionuclides of $^{226}$Ra, $^{232}$Th, and $^{40}$K in the collected samples is calculated using the following formula (Mamont-Ciesla, et al., 1982).

$$AGDE (\mu Sv.y^{-1}) = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_{K}$$  \hspace{1cm} (6)

III. RESULTS AND DISCUSSION

Fig. 4 shows the activity concentrations in (Bq.kg-1) of the radionuclide $^{137}$Cs and $^{226}$Ra, $^{232}$Th, and $^{40}$K for 36 investigated wheat grain samples. The radionuclide $^{137}$Cs was not detected in the all measured wheat grain samples. The activity concentration of $^{226}$Ra varies from 0.245 ± 0.116 Bq.kg$^{-1}$ (WS$_{10}$ - Ella Allah village) to 0.746 ± 0.086 Bq.kg$^{-1}$ (WS$_{15}$ - Kani lala village) with an average value of 0.407 ± 0.097 Bq.kg$^{-1}$. The activity concentration of $^{232}$Th was below minimum detectable activity in the samples of WS$_{5}$, WS$_{5}$, WS$_{6}$, WS$_{22}$, WS$_{27}$, WS$_{29}$, WS$_{37}$, and WS$_{55}$ and it was not detected of two samples (WS$_{1}$ and WS$_{34}$). This activity ranged from below minimum detectable activity BMDA to 0.814 ± 0.367 Bq.kg$^{-1}$ (WS$_{16}$ - Talabani gawra) with an average value of 0.36 ± 0.14 Bq.kg$^{-1}$. Furthermore, the activity concentration of $^{40}$K was found in all samples with the minimum value of 72.04 ± 1.561 Bq.kg$^{-1}$ (WS$_{1}$ – Pebazok) and the maximum value of 136.1 ± 2.659 Bq.kg$^{-1}$ (WS$_{31}$ – Sinawa), with average value of 109.25 ± 2.214 Bq.kg$^{-1}$. The activity concentration of $^{40}$K has been high because it is naturally high abundance in environmental samples. Moreover, $^{226}$Ra detection in wheat grain samples was expected, because it is a daughter product in the decay series of $^{238}$U which is typically found in environmental samples. Moreover, the activity concentration of $^{232}$Th was not detected or BMDA below minimum detectable activity in some wheat grain samples, but it does not imply absolutely that the absence of $^{232}$Th in these samples. In fact, many researchers in their studies have reported BMDA or non-detection for $^{232}$Th in wheat grains (Changizi et al., 2013, Abojassim et al., 2015, Hosseini et al., 2006). The obtained results showed the activity concentrations of the radionuclides ($^{226}$Ra $<^{232}$Th $<^{40}$K) which is in accordance with the information presented by (Changizi et al., 2013). The soil-to-wheat grain transfer factors of $^{40}$K are considerably higher than those for $^{226}$Ra and $^{232}$Th because of the high solubility of $^{40}$K in water and its high mobility in soil (Kumar, et al., 2008). The noticeably high recorded values of $^{40}$K in the wheat grain samples within the present study are similar findings recorded by Akhtar and Tufail, 2006, Alshahri, 2016. The average values of activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K for the wheat grain samples in this study were too lower than the worldwide average values recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation Sources as 32 Bq/kg for $^{226}$Ra, 45 Bq/kg for $^{232}$Th, and 412 Bq/kg for $^{40}$K (UNSCEAR, 2000).

Furthermore, Fig. 4 shows the variations of concentration levels of $^{226}$Ra, $^{232}$Th, and $^{40}$K according to the different wheat grain samples. These variations may be due to the different concentration of the radionuclides $^{226}$Ra, $^{232}$Th, and $^{40}$K in the soils of wheat plantation fields which could be absorbed by wheat plants (El-Taher and Makhluf, 2010). Overall, the obtained results indicated that radioactivity levels in the
wheat grain samples collected from the wheat plantation fields of Koya district are not at the range of health risk.

Furthermore, there is study in Iraq, aiming at clarifying the radiation hazard indices and ingestion effective dose in wheat flour samples collected from Iraqi markets was conducted by (Abojassim et al., 2015), by used NaI (Tl) detector was used to radiometric analysis for 12 different types of flours those were available in Iraqi markets. The specific activities were varied from 1.086 ± 0.0866 to 12.532 ± 2.026 for \(^{238}\)U, from 0.126 ± 0.066 to 4.298 ± 0.388 for \(^{232}\)Th, and from 41.842 ± 5.875 to 264.729 ± 3.843 for \(^{40}\)K. The average values of radium equivalent and internal hazard index in wheat flour samples were found to be 19.6347 Bq.kg\(^{-1}\) and 0.0708, respectively.

Comparisons between the natural radioactivity levels in wheat grain samples of the present study with some other studies among worldwide listed in Table I.

Table II shows the values of radium equivalent activity \(R_{\text{eq}}\) of the wheat grain samples. The results of \(R_{\text{eq}}\) for the measured wheat grain samples were ranged from 6.71 to 11.9 Bq.kg\(^{-1}\) with average value of 9.33 Bq.kg\(^{-1}\) that is less than the permissible limit (370 Bq.kg\(^{-1}\)) (UNSCEAR, 2000), this result indicates that the collected wheat grain samples among the wheat farming lands of Koya district have no radiation hazards. The values of other parameters such as internal and external hazard indices \(H_{\text{in}}\) and \(H_{\text{ex}}\) and AGDE due to the specific activities of \(^{226}\)Ra, \(^{232}\)Th, and \(^{40}\)K are presented in Table II, the values of \(H_{\text{in}}\) and \(H_{\text{ex}}\), and AGDE were ranged from 0.019 to 0.03 with average value of 0.026, from 0.018 to 0.032 with average value of 0.025, and from 26.1 \(\mu\)Sv.y\(^{-1}\) to 46.97 \(\mu\)Sv.y\(^{-1}\) with average value of 37.06 \(\mu\)Sv.y\(^{-1}\), respectively, the lowest values of \(H_{\text{in}}\) and \(H_{\text{ex}}\) of the wheat grain sample were found in Pebazok village. Whereas, the highest values of the wheat grain sample were found in Siktan village. The obtained results were compared to the recommended permissible limits. This study indicated that the average values of \(H_{\text{in}}\) and \(H_{\text{ex}}\) of wheat grain samples were found to be lower than unity (<1), this reveals that the radiation hazards due to the wheat grain samples among the
studied area are insignificant. Moreover, the average value of AGDE was lower than the permitted limit of 300 µSv.y⁻¹ as given by UNSCEAR, 2000.

The annual effective ingestion dose due to the consumption of wheat grains was calculated based on annual intake of 134.5 kg.y⁻¹ (dry weight) of wheat grains by adults in Iraq, Kurdistan region, as given by Azeez, et al., 2019. The ingestion dose due to the intake of each of natural radionuclides ²⁲⁸Ra, ²³¹Th, and ⁴⁰K is presented in Table III. The calculated values were ranged from 7.02 to 20.06 µSv.y⁻¹ with the average value of 10.95 µSv.y⁻¹ of E_{ING} (²⁸Ra), from 0 to 25.18 µSv.y⁻¹ with average value of 11.14 µSv.y⁻¹ of E_{ING} (²³¹Th), and from 60.07 to 113.49 µSv.y⁻¹ with a mean value of 91.1 µSv.y⁻¹ of E_{ING} (⁴⁰K). The total ingestion dose E_{ING} (total) due to the summation of E_{ING} (²⁸Ra), E_{ING} (²³¹Th), and E_{ING} (⁴⁰K) ranges from 80.7 to 141.59 µSv.y⁻¹ with average value of 113.19 µSv.y⁻¹ which is twice smaller than the worldwide average value of 260 µSv.y⁻¹ as recommended by UNSCEAR, 2000. Radionuclide absorption from soil by plants depends on the soil characteristics which include pH content, clay content, soil texture, cation exchange capacity, dominant clay minerals, exchangeable cations, and organic matter content. In addition, the uptake of radionuclides is affected by the plant type and type of radionuclides – the radionuclide is heavy or light element (Konoplev, et al., 1993). Thus, this study revealed that the radiation hazard due to the total internal dose by the intake of ²⁸⁸Ra, ²³¹Th, and ⁴⁰K of the consumption of wheat grains is insignificant. In Fig. 5, the results show that the average ingestion dose due to the intake of ⁴⁰K (91.1 µSv.y⁻¹) is more than average ingestion dose for both ²²⁸Ra (11.14 µSv.y⁻¹) and ²³¹Th (10.95 µSv.y⁻¹).
but the results value of average ingestion dose ingestion dose for $^{226}\text{Ra}$ and $^{232}\text{Th}$ is so near together because the half-life for both have patents is very long.

IV. CONCLUSIONS

This research aimed to measure the natural radioactivity levels wheat grain samples from the wheat plantation fields of Koya district, the average of concentration of radionuclides of $^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$ in the wheat grain samples was found to be lower than the worldwide average values recommended by UNSCEAR, but no detection of $^{137}\text{Cs}$ in the wheat grain samples. The total annual ingestion dose due to the intake of natural radionuclides of $^{226}\text{Ra}$, $^{232}\text{Th}$, and $^{40}\text{K}$ by the consumption of wheat grains was equal half of the world average value of 260 $\mu$Sv.y$^{-1}$ as given by UNSCEAR, therefore, the accumulation of primordial radionuclides in the wheat grains was produced from the wheat plantation fields of Koya district does not have a significant health risk. The obtained results of the this study would be a useful data for making a baseline of artificial and natural radioactivity and heavy metal concentration levels in soils of the studied area. These baseline data will help us to assess any variations in the radioactivity levels due to any unexpected events such as nuclear reactor accidents and/or nuclear weapon tests or due to the anthropogenic activities within the study area.

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