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Original article

Detection of Types and Contents of Natural Radionuclides and Hazard Indexes in Vegetable and Soil Samples at Some Al-Marj City Locations

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Abstract
This study aims to evaluate the natural radioactivity concentrations of naturally occurring in the environment associated with the ^{26}Ra ^{238}U, and ^{232}Th decay chains and the long lived natural occurring radionuclide ^{40}K in vegetables and soil samples. The samples were collected from different locations around Al MARJ region (north eastern side of Libya) to elevate natural radioactivity. The analysis of the samples was done by using Sodium iodide detectors. The analysis demonstrates that the measured activity concentrations for the soil samples of ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K were (60.47 to35.37), (51.31 to 37.29), (68.94 to 28.43), and (271.36 to 53.72) Bq/Kg, respectively. While the average activity concentrations for vegetable samples of ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K were (86.06 to 32.37), (91.85 to 38.87), (114.26 to 60.60), and (280.41 to 66.92) Bq/kg, respectively. The radium equivalent activity (Raeq) in the soil and vegetable samples was calculated, and the results recorded that the values ranged between 132.79 and 87.13 Bq/kg, and 227.18 to 124.19 Bq/kg respectively. The values of external hazard index, gamma radiation level index (Iγ), and I-alpha (Iα) are close to unity for all samples, which was investigated. The absorbed dose rate, annual effective dose, and cancer risk factor were determined for all samples. The study concluded that the highest activity concentration in soil and vegetable samples is higher than the worldwide average as reported by the UNSCEAR.
Keywords. *Radioactive Nuclides, Vegetable, Soil, Libya.*

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Introduction

Radionuclides are found throughout natural and it exists in the soil and vegetables. These radionuclides have half-lives that are approximately Earth's age or older (i.e., about 4 to 5 billion years) [1]. Natural radioactive decay series such as ^{238}U and ^{232}Th as well as singly occurring radionuclides such as 40K exist in the earth and atmosphere in varied levels. The radioactivity present on air or in the agricultural land and in soil may transfer to the crops grown on it. It happens, however, that an amount of some radioactive elements finds their way into human bodies [2]. Generally, the plants (vegetables) may cause accumulation of radionuclides in their organs, which may additionally rely on the chemical and physical properties of the soil. So, there may be multiplied risk to human population via food chain. The main sources of components from the environment to plants are: air, water and also the soil [3].

There are two ways for transferring of the radionuclides present in the environment in to plants by indirect and direct methods. The first method (indirect) happens by uptake from soil through roots. When plants are grown in the contaminated soil, the radioactivity is transferred from the soil to the roots and then in shoots plant. In the end, the radioactivity is shifted to the human diet. These radionuclides will get transferred into plants together with the nutrients throughout mineral uptake and accumulation in varied components and even reach edible portions [4]. The second method (direct) happens by absorption through aerial elements of the plants. Presence of emission (alpha, beta and gamma) in plant organs. Naturally the radioactive contamination of the soil affects the plants, animals and human beings living in the given area directly as the radioactive isotopes accumulated in the soil, cause direct radiation exposure [5].

Soil and vegetables are recognized as one of the major pathways for the transfer of radionuclides to human beings. The study of natural radioactive elements was determined in samples of fishes and soils at different locations of Libya, most of these studies recorded high values of these elements [5-7]. The measuring of hazard compounds took place in different samples of many studies [8-56]. The main aims of this study was to Detect and Estimate the types and quintiles of Natural radioactive nuclides in some vegetables and soil samples collected from Al-MARJ region, Libya.

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Methods

Description of the Studied Area

The area of study is located in the northern east of Libya at (ALMarj region, Libya) which has a semi-humid environment with an average annual rainfall of around 382 mm/year. The farms are spread out over a large region. The zone has been heavily impacted by pollutants from agricultural chemicals and some human activities. The soil classified under the Rendolls class, which is rich in calcium carbonate and is immature, with no deep profile on the prospects for well-known for farmers' inclination toward irrigated agriculture, which has prompted the increased use of fertilizers to boost soil fertility, as well as the increased use of chemical pesticides for pest control, which has expanded owing to the growth of irrigated agriculture. Ten Samples locations were selected in this study including: (line 16 farms, Old ALMarj, New Al-Marj, Almhaadi, Farzogha- A gsenta -Zawet ALgosor- Awiliyah- Al-Ahmadah and AL slayaya), Figure 1.

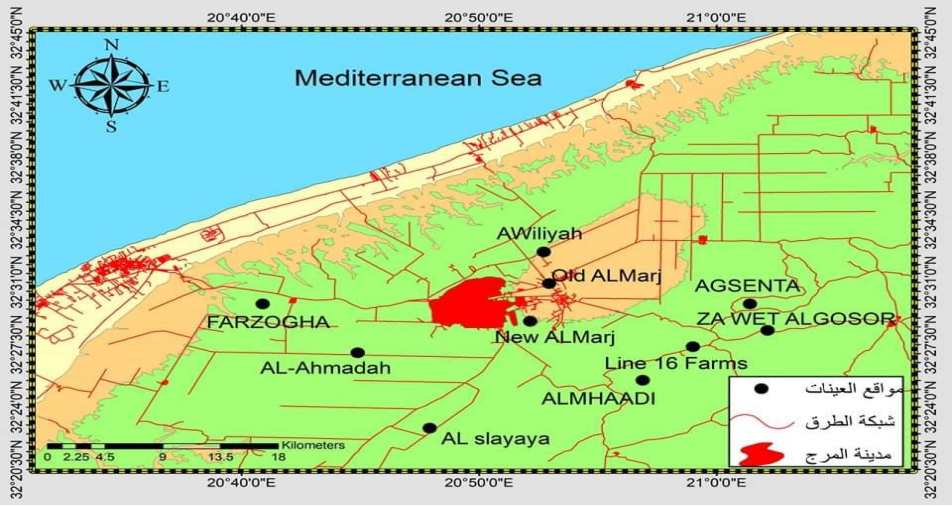

Figure 1. The study locations.

Sampling and Preparation of Samples

Vegetable Samples

The vegetable samples were collected randomly from several locations at some farms, During (2021) which covering about 100 km². Five species of vegetable types (Cucumber, Zucchini, Bean, Cabbage and Peas) selected in this study. These vegetable samples were product in five locations as shown in Table (1) Approximately (2 kg) of each vegetable samples were taken; the studied samples are shown in the Table (1). Also, ten locations were selected for soil samples, where the samples collected from the locations of vegetable farms in addition to (samples) collected the area around Al-Marj region which not product vegetables at the sampling time. (1Kg) of surface soil samples were collected each farm. Both soil and vegetable samples were protected in plastic bags and transported to the laboratory. Fresh samples were washed with tap water, then by deionized water, air dried and then carefully weighed. The samples were dried after chopped into small pieces. Then dried again in oven at 75 °C for 36 hours, grinded to homogeneous small pieces by using food processor (blender), then grinded to powder and sieved through a 2 mm nylon sieve to obtain a representative sample. and appropriate volume of sample was transferred into specimen cans.

Table 1. Vegetable sample's locations:

Vegetable	Location
Cucumber	New ALMarj
Zucchini	Awiliyah
Bean	Old ALMarj
Cabbage	Al-Ahmadah
Peas	Farzogha

Collection and preparation of the soil samples

Soil samples (about 1 Kg) were collected and transferred into a clean polyethylene bag from the same sites of the vegetable samples (for each vegetable type separately), where ten samples were taken from 0 – 20 cm depth using a

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steeliness steel auger and pooled together to form composite sample. The collected soil samples were transported to central laboratory of chemistry at faculty of science (Omar Al-Mukhtar University). The soil samples were air dried in a dry and dust free place at a room temperature of (25 °C) for 5 days, followed by an oven dry at 100 ^{0}C for 24 h. The samples were grinded with a pestle to pass through a 2 mm sieve and homogenized. The dried, sieved and homogenized soil samples were finally stored in polyethylene bags and kept in desiccators until digestion and analysis [6-7].

Radioactive elements:

The natural radioactive nuclei were determined by Sodium Iodide (NaI) detector method.

Preparation of Samples of NaI Detector:

Before measured samples by sodium Iodide soil and vegetable samples were dried at the temperature of 95C^{o} for 3 and 4 hours until the moisture was completely removed. The samples were then smashed into fine particles and thoroughly mixed, and passed through a fine mesh sieve (~200 mesh) to obtain composite representative samples. After that weighted the samples are placed in polyethylene bottles of 250 cm^{3} volume. The bottles were kept airtight for a month before being putting in to NaI spectrometer, in order to secure the secular equilibrium between long-lived radioisotopes ^{238}U, ^{232}Th, and their corresponding daughters. This step is very necessary to ensur the radon gas is confined within the volume and the daughter still also remain in the sample. These samples were placed directly over the detector. The counting time for each sample was 70000 sec. The measured activity concentrations were presented as Bq/kg. Also some radioactivity indexes were calculated according to the following methods [6-7]:

Radium Equivalent Dose (Ra_{eq})

The radium equivalent **(Bq./kg)** is estimated by weighted sum of activities for radionuclides of ^{226}Ra,^{232}Th, and ^{40}K in sample and it is given by the relation:

$$Ra_{eq} = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_{K}$$

Where:

A_{Ra}, A_{Th} and A_{K} are the activity concentration (Bq./kg) for^{238}U,^{232}Th, and ^{40}K respectively [5].

External Hazard Index (H_{ex}):

The external hazard index (H_{ex}) due to emitted from gamma ray of the samples to estimate the biological hazard was calculated by the relation:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \leq 1$$

Where: A_{Ra}, A_{Th} and A_{K} are the activity concentrations for ^{226}Ra,^{232}Th, and ^{40}K in Bq/kg .

Internal Hazard Index (H_{in}):

Internal exposure arises from the inhalation of radon (^{222}Rn) gas and its progeny products or ingestion of other radionuclides. Since radon is carcinogenic, it is present in all building materials. Hence for the measurement of radon exposure, the internal hazard index is given as following (Al-Harbi and El –Taher, 2016):

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

Radiation Level Index (I γ) :

The radiation level index I γ is used to assessment the hazard level of radionuclides 238U,^{232}Th, and ^{40}K. The radiation level index calculated by using the relation:

$$I_{\gamma} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_{K}}{1500}$$

Where: A_{Ra}, A_{Th} and A_{K} are the activity concentrations for ^{226}Ra,^{232}Th, and ^{40}K in Bq/kg [7].

Alpha index (Iα):

The external irradiation, especially radon and its short-lived products are also hazardous to respiratory organs, they emit radioactive alpha particles and attach to aerosols, dust and other particles in the air. As we inhale, radon progeny are deposited on the cells lining the airways where the alpha particles can damage DNA and potentially cause lung cancer.

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The excess alpha radiation due to radon inhalation is estimated from the alpha index (Iα), which is defined as following:

$$I_\alpha = \frac{A_{Ra}}{200}$$

The recommended upper limit concentration of ^{226}Ra is 200 Bq/kg which gives Iα = 1.

Description of The System:

Sodium iodide scintillation NaI(Tl) detectors using to measure the samples. The detector was insert in the cylindrical lead shield to reduce the background and noise radiation from many natural radionuclides, such as, (^{40}K), decay series, and cosmic rays. The based system of gamma ray spectroscopy consists of a NaI (Tl) detector, high voltage power supply, Multi-Channel Analyzer (MCA box) and Sensor Cassy. The gamma ray spectra data was analyzed by using Cassy Lab software on a PC.

Results

For the analysis of samples in the (soil and vegetables), the background measurement is very important. The background radiation spectrum of the ionized water is obtained for two days (14,0000 sec) and the other samples for one day (70000 sec). Different sources contribute to background radiation around the detection environment. This includes ambient air for the detector, shielding and electronic components of the detector itself.

Result of radioactive nuclides from NaI detector Spectrometer:

For all samples the activity concentration of ^{226}Ra determined from photo peak 186.2 KeV. The activity concentration of ^{238}U series determined ^{214} Pb photo peak (295.2 and 351.9) KeV and ^{214}Bi (609.3, 768.4, 1120.3, 1238.1, 1377.7 and 1764.5) KeV. Similarly, for ^{232}Th series, the activity concentration determined from ^{228}Ac (92, 209.5, 338.5, 911.1 and 968.9) KeV and ^{208} Tl 583 KeV ^{212}Bi 727.2 KeV. ^{40}K was determined from photo peak 1460 KeV [5-7].

For soil samples the activities of ^{226}Ra, ^{238} U, ^{322}Th and ^{40}K were ranged between (35.37- 60.47), (37.29 – 51.31), (28.43 - 68.94) and (53.72 – 271.36) BqKg^{-1}, for ^{238} U, ^{322}Th, ^{226}Ra, and ^{40}K, respectively. The minimum activities in soil samples of ^{226}Ra, ^{238} U, ^{322}Th and ^{40}K were 35.37, 37.29, 28.43 and 53.72 BqKg^{-1}, respectively and the maximum values were 60.47, 51.31, 68.94 and 271.36 BqKg^{-1}, respectively. The activity concentrations in the vegetable samples were fluctuated in the ranges of (32.37 -86.06), (38.87 – 91.85), (60.60 – 114.26) and (66.92 – 280.41 Bq/Kg) for ^{226}Ra, ^{238} U, ^{322}Th and ^{40}K respectively. For the most samples the activity concentrations of ^{226}Ra for soil, vegetable samples under investigation are higher than those the world's population with average of (32) Bq/Kg(UNSCEAR, 2008), respectively. For 238U the activity concentrations of all samples are higher than the permissible level for all samples under investigation (33 and 20) Bq/Kgand 1 BqL^{-1} [4]

The average activity concentrations of radionuclides (^{232}Th) in soil and vegetable samples (45, 15 Bq/Kg and 0.05 BqL^{-1} for ^{232}Th) are higher than the permissible level ,but for ^{40}K the activity are lower than the permissible limits (412, 420 Bq/Kg and 10 BqL^{-1} for ^{40}K) Table (2) and Figures(1-4).

Table 2. The activity concentrations (Bq/kg) of the radionuclides ^{226}Ra,^{238}U, ^{23} Th and ^{40} K of the investigated samples.

Samples Code	Ra^{226}	^{238}U	^{232}Th	^{40}K
S1	35.37	37.29	36.66	53.72
S2	44.00	45.99	39.29	271.36
S3	36.39	42.91	59.06	122.25
S4	46.28	42.75	57.82	246.57
S5	52.99	51.31	29.93	104.03
S6	37.21	38.95	28.43	120.42
S7	60.47	44.31	68.94	98.58
S8	51.76	49.29	42.89	165.50
S9	45.70	39.10	28.76	64.80
S10	56.65	47.98	44.52	162.00
P.L	32	33	45	412
V11	44.41	50.64	64.69	280.41
V12	32.37	38.87	60.60	66.92

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V13	53.75	55.27	114.26	123.71
V14	86.06	91.85	85.29	248.74
V15	57.01	54.47	84.06	150.55
P.L	**50**	**20**	**15**	**420**

P.L = Permissible level

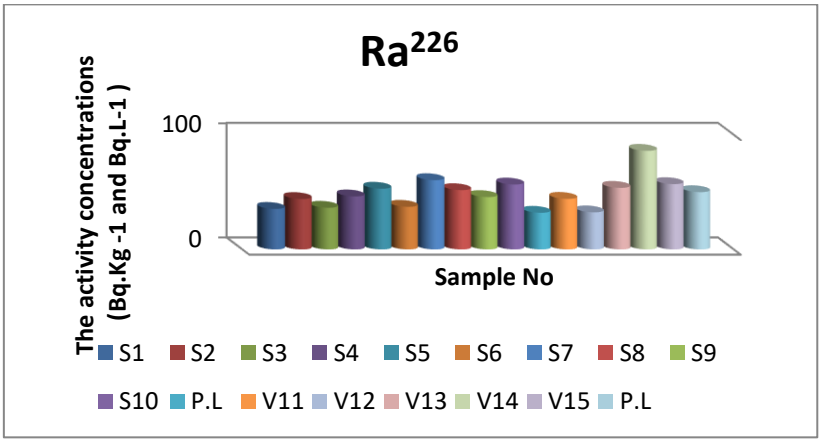

Figure 1. Comparative of the contents of ^{226}Ra activity between the soil and vegetable samples.

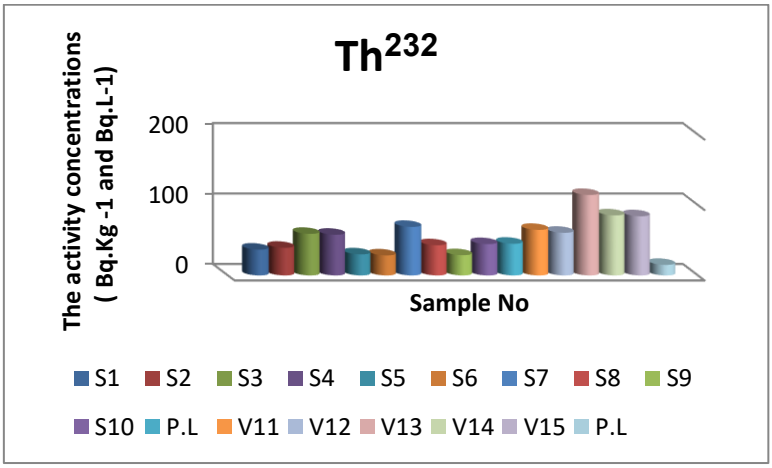

Figure 2. Comparative of the contents of ^{232}Th activity between the soil and vegetable samples.

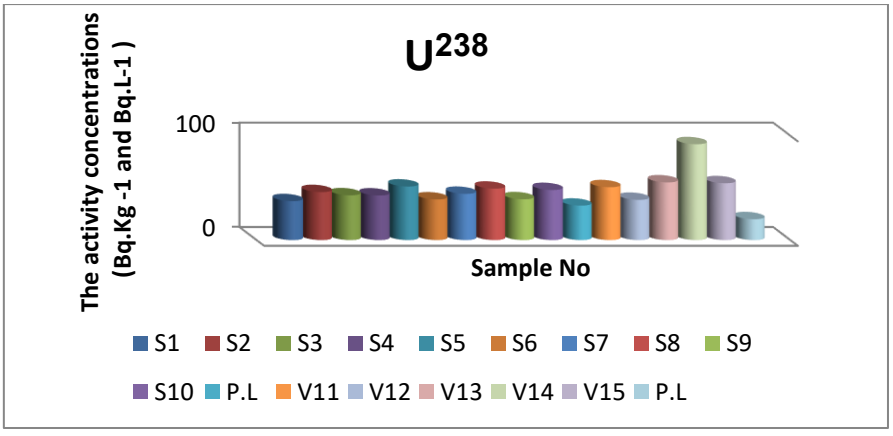

Figure 3. Comparative of the contents of ^{238}U activity between the soil and vegetable samples.

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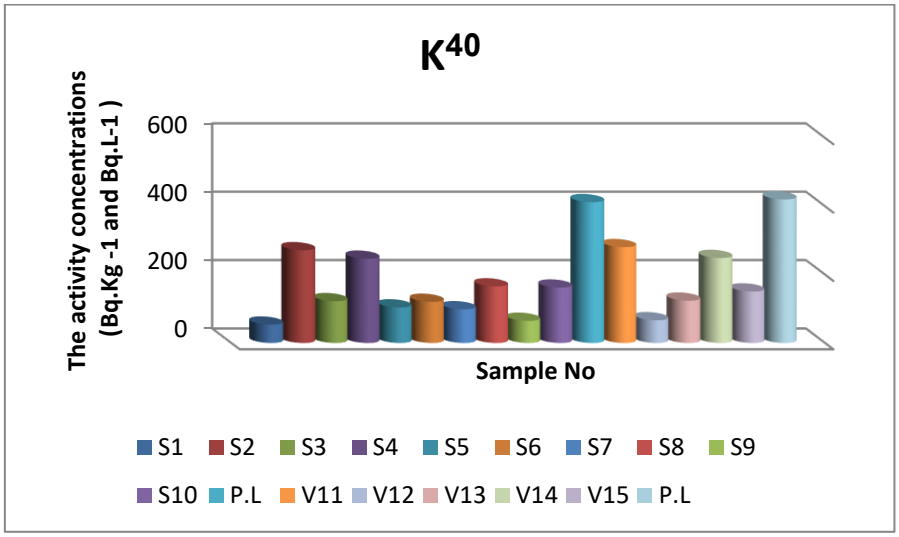

Figure 4. Comparative of the contents of ^{40}K activity between the soil and vegetable samples.

Radium Equivalent (Ra_{eq})

The radium equivalent values of all samples were presented in Table (3), for the soil samples the values were ranged (87.13 – 132.79) BqKg.$^{-1}$ The radium equivalent for vegetable samples were ranged between (124.19 – 227.18) BqKg.$^{-1}$, Figure (5). The radium equivalent values for all samples are lower than the permissible limit level of (370 BqKg.$^{-1}$),[4].

Internal Hazard (H_{in})

The internal hazard values obtained in Table (2) and Figure (6). The internal hazard of soil samples in ranged from (0.34 – 0.61). The external hazard values of vegetable samples were ranged between (0.42-0.85) mGy.y.$^{-1}$ The internal hazard values for all samples are lower than unity of permissible level of (1),[4].

External Hazard (H_{ex})

The external hazard values obtained in Table (3) and Figure (6). The external hazard of soil samples in ranged from (0.23 – 0.40). The external hazard values of vegetable samples were ranged between (0.34- 0.61) mGy.y.$^{-1}$ The external hazard values for all samples are lower than unity of permissible level of (1),[4].

External Gamma Radiation ($I\gamma$)

The calculated **$I\gamma$** values for all samples were shown in Table (3) and Figure (5&6). The values of soil samples were ranged from (0.61 – 1.16). The calculated **$I\gamma$** values of vegetable samples were ranged between (0.87- 1.59) The calculated values of external gamma radiation for most soil and vegetable samples were higher than the international values (**$I\gamma$=1**).

Alpha Index

The values of **$I\alpha$** calculated from the concentrations of ^{226}Ra are presented in Table (3) and Figure (5&6). However, the alpha index in soil samples has been found to vary from ranged (0.18 – 0.30). In addition for the vegetable samples the values ranged (0.16 – 0.43) . It is observed that all values of **$I\alpha$** are below the maximum permissible value of (**$I\alpha$=1**).

Excess Lifetime Cancer Risk (ELCR):

Table (3), shown the cancer risk factor **ELCR** for all samples. The values of soil samples obtained were ranged from (0.15-0.29). The values for vegetable samples were ranged between (0.21 – 0.40). The values for some samples are higher than the international values of 0.29×10^{-3} .

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Table 3. The value s of radium equivalent, internal hazard, external hazard, gamma index and alpha index.

Samples code	Ra_{eq}	H_{in}	H_{ex}	$I\gamma$	$I\alpha$	ELCR $\times 10^{-3}$
S1	91.93	0.34	0.25	0.64	0.18	**0.16**
S2	121.08	0.45	0.33	0.87	0.22	**0.22**
S3	130.25	0.45	0.35	0.91	0.18	**0.23**
S4	147.96	0.52	0.40	1.05	0.23	**0.26**
S5	103.80	0.42	0.28	0.72	0.26	**0.18**
S6	87.13	0.34	0.23	0.61	0.19	**0.15**
S7	166.65	0.61	0.45	1.16	0.30	**0.29**
S8	125.84	0.48	0.34	0.88	0.26	**0.22**
S9	91.82	0.37	0.25	0.63	0.23	**0.16**
S10	132.79	0.51	0.36	0.93	0.28	**0.23**
V11	158.51	0.55	0.43	1.13	0.22	**0.28**
V12	124.19	0.42	0.34	0.87	0.16	**0.21**
V13	226.68	0.76	0.61	1.58	0.27	**0.39**
V14	227.18	0.85	0.61	1.59	0.43	**0.40**
V15	188.81	0.66	0.51	1.32	0.28	**0.33**
P.L	**370**	**1**	**1**	**1**	**1**	**0.29**

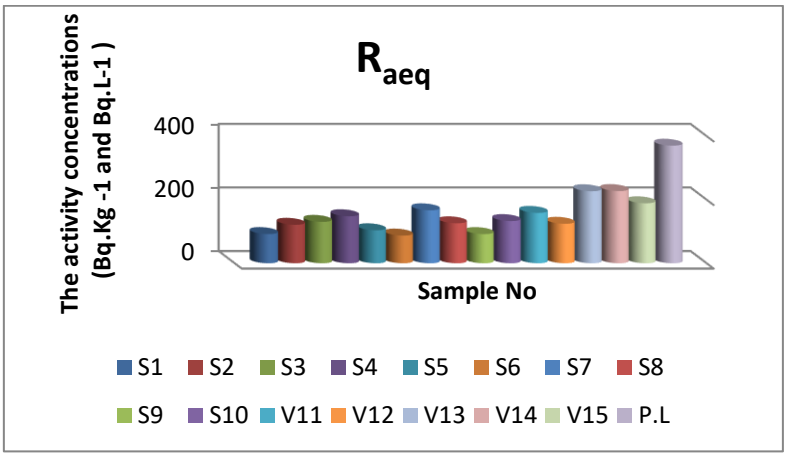

Figure 5.The values of Ra $_{eq}$ of the studied samples.

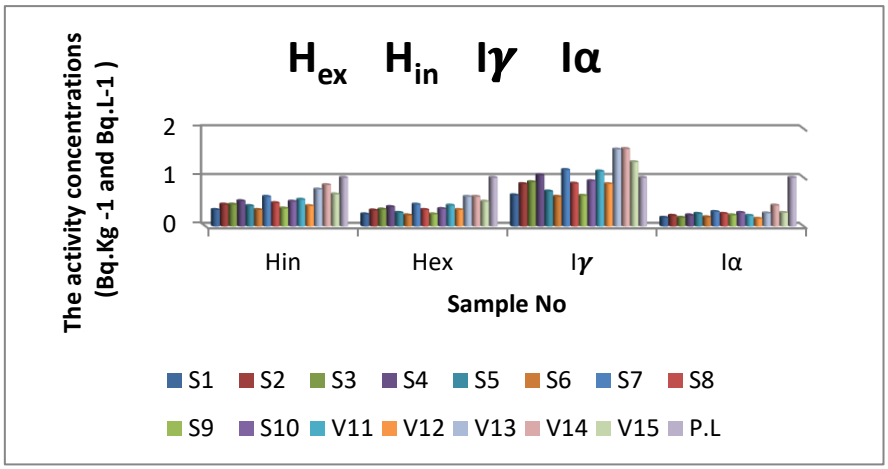

Figure 6. The values of Hex, Hin, Iγ and Iα of the studied samples.

Discussion

It was reported that some of the toxic elements interact with some compounds to produce many new compounds that have different medical applications [57-67]. In Libya, many studies were carried out on different samples to evaluate

the contents of hazardous materials [68-100]. In this study, the results showed that the contents of radioactive nuclides were varied between the studied samples. Uranium, thorium, and radium are radioactive elements that originate naturally in the soil. Uranium is the most abundant in the earth's crust and also tends to spread during the soil because the rocks in the outer crust have suffered from weathering processes (water, air, plants) as a result of these processes and different factors other the soil is formed, the soil pollution depends on relevant information transport and disposal operations away from the site of contamination, Because the accumulation of radioactive material and its movement depends on the interaction of materials and compounds with the hard part of the soil and the type of this reaction reflects the soil's ability to retain radioactive materials and on the other hand the rate of fall rainfall, quantity of irrigation water, type of cultivated plant and soil management processes lead to the movement of radioactive contaminants into groundwater or their transfer to plants or other medium such as water and air, radioactivity includes soil models of radionuclides, which belong to the 238U series and the most important of radioactive element is ^{226}Ra,[101].

Radiation pollution in groundwater is due to radium resulting from the melting of underground reservoir rocks, and 222Ra, which is highly soluble in water. There are radionuclides produced by the decomposition of uranium, thorium, granitic and sedimentary rocks, which produce radioactive materials. Sedimentary rocks form the late uranium-rich Cretaceous rocks, which are spread over a wide area of North Africa and the Middle East. The high concentrations of uranium, radon, and radium in drinking water cause genetic mutations, deformed births, and serious cancerous diseases. Most of them need hundreds of years to have no negative impact on human life. The topography of the topsoil of the aquifer has a large role in hiding and increasing pollution in groundwater [102]. The soil's own ability to purify polluted water is related to soil quality and its properties that allow or impede the movement of pollutants into groundwater. There are highly purified soils that contribute significantly to water purification from their pollutant before they reach ability to groundwater [102].

The higher average levels of ^{238}U than average global values in the studied area may be due to the use of phosphate fertilizers required to treat soil depletion of natural nutrients removed through agriculture and erosion. This fertilizer is composed of phosphate rocks that contain a high concentration of uranium. Elevated levels of ^{232}Th indicate the presence of carbonate and monazite rocks, which are known to be rich in these radionuclides. The high levels of ^{40}K are due to the fact that the soil samples were collected from agricultural land where there is application of some potassium-rich inorganic fertilizers and other chemicals used to promote crop production. The variations of radioactivity elements in soil samples depend primarily on the geological structure and geographical conditions, such as the location of the study area, which can significantly contribute to the presence of radioactive elements. Therefore, the high percentages of these nuclei in the soil also affect the concentrations in vegetable samples. The concentrations of radionuclides of ^{226}Ra, ^{238}U, and ^{232}Th (ppm) and ^{40}K for all samples at the Almarj area. The use of some Fertilizers s and pesticides may be affecting the distribution of radioactivity, where there is a relative increase in the contents of vegetables and soils collected from the same farms.

Conclusion

According to the results recorded in this study, the levels of radioactive nuclides were detected in all samples as: 238U, ^{40}K, ^{232}Th, and ^{40}Ra. This study would help to reduce the risk to human health that originates from highway pollution. But further extensive sampling is necessary to include similar study areas, and further research concerning contamination of environmental.

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Conflict of interest. Nil

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