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Evaluation of radioactive contamination in soil samples from the Diyala Bridge area, Baghdad/Iraq: gamma spectroscopy and radon inhalation dose estimation

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Abstract

This study evaluates the specific activity levels of Ra-226 (186 keV), Bi-214 (609 keV), Cs-137 (662 keV), Ac-228 (969 keV), and K-40 (1460 keV) in soil samples from the Diyala Bridge/Baghdad area using a NaI(Tl) scintillation detector for gamma-ray spectroscopy. Key parameters calculated include Radium Equivalent Activity, Gamma Dose Rate, External and Internal Hazard Indices, and the Gamma Index. In the results, the calculated Excess Lifetime Cancer Rate due to exposure to gamma ray were varied from 0.460×10^{-6} in sample (9) to 2.721×10^{-6} in sample (5), with an average of 1.236×10^{-6} across all samples. Also in this paper the concentration of Rn-222 in the air was estimated based on radium data, and doses from radon gas consumption and inhalation were calculated. The average inhalation dose from radon in soil samples was 47.941×10^{-9} Sv/y. All radionuclide activity levels were below permissible limits except in sample (5), located near the Iraqi Atomic Energy facility in Al-Twetha city.

Keywords: NaI(Tl) scintillation detector, Soil samples, Radon gas, Diffusion rate.

تقييم التلوث الإشعاعي في عينات التربة من منطقة جسر ديالى، بغداد/العراق: التحليل الطيفي لأشعة كاما وتقدير جرعة استنشاق الرادون

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الخلاصة

تقدم هذه الدراسة تقييم مستويات النشاط النووي الخاصة لكل من **Ra-226 (186 keV)** و **Bi-214 (609 keV)** و **Cs-137 (662 keV)** و **Ac-228 (969 keV)** و **K-40 (1460keV)** في عينات تربة من منطقة ديالى جسر/ بغداد بطريقة التحليل الطيفي لأشعة كاما باستخدام الكاشف الوميضي **NaI(Tl)**. بعد ذلك، تم تحديد معاملات مثل النشاط المكافئ للراديوم، ومعدل امتصاص الجرعة الإشعاعية ومؤشر الخطر الخارجي والداخلي ومؤشر الجاما التمثيلي. وتراوح معدل الإصابة بسرطان الحياة الزائدة من 0.460×10^{-6} للعينة (9) إلى أعلى قيمة تبلغ 2.721×10^{-6} للعينة (5)، بمتوسط يبلغ 1.236×10^{-6} عبر جميع العينات. بالإضافة إلى ذلك، تم تقدير تركيز **Rn-222** في الهواء استنادًا إلى بيانات تركيز الراديوم، وتم حساب الجرعات الناتجة عن تناول واستنشاق غاز الرادون باستخدام المعادلات الرياضية ذات

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الصلة. وتبين أن المتوسط المتوقع للجرعة الناتجة عن استنشاق غاز الرادون في عينات التربة هو 47.941×10^{-9} سيفرت/سنة. ولاحظ أن مستويات النشاط النووي الخاصة لجميع النواتج الإشعاعية كانت دون الحدود المسموح بها باستثناء العينة (5)، والتي كانت على مقربة من منظمة الطاقة الذرية العراقية في مدينة التويثة.

1. Introduction

During the twentieth century, significant technological advancements aimed at benefiting humanity were achieved. However, these advancements also brought about considerable risks that directly impacted the environment, thereby affecting human health and societal well-being. These technologies include radioisotopes that emit radiation that can cause both immediate and long-term harm to humans and the environment [1]. Humans are permanently exposed to ionizing radiation from two main sources: natural and man-made sources that are made for various purposes. Exposure to natural sources, such as cosmic rays and rays emanating from naturally radioactive elements, constitutes the main percentage of exposure. Naturally occurring terrestrial radionuclides primarily stem from primordial radionuclides with significantly long half-lives [4]. Almost all materials surrounding us contain small numbers of radioactive materials. Therefore, humans are exposed to a low level of background radiation, which affects the environment, and the effects may persist for several years. It may affect the genetic makeup of humans and animals, leading to a genetic defect that will appear in future generations.

The quality of soil depends heavily on human interaction and its usage. Soil serves as a crucial resource that provides the necessary environment for plant growth, supplying essential minerals and nutrients that sustain human food production. It consists of a complex three-phase system comprising solids, liquids, and gases [2]. Defined as natural bodies found on the Earth's surface, soil contains living matter and supports, or has the potential to support, plant life [3]. Its composition is highly variable in terms of physical and chemical attributes, containing organic and inorganic compounds critical for plant growth.

Several studies are concerned with measuring the specific activity of natural radioactivity in the soil. Al-Hayani et al. studied the concentration of natural radionuclides in the soil of Salah Al-Din Governorate using gamma spectroscopy with NaI(Tl) detector [5]. Shihab et al. studied the concentrations of radon and natural radionuclides in soil oil wells in (Majnoon), Basrah using gamma spectroscopy with NaI(Tl) detector [6]. In 2022, Siyah et al. studied the concentration of natural radionuclides in the soil of the Al-Nahrawan site using a high-purity germanium (HPGe) detector [7]. Moreover, the effect of these radionuclides in water was studied [8]. In addition to the presence of radionuclides in the soil, radon, a member of the radioactive chain of uranium, is emitted from soil and enters the food chain of humans and animals [9]. Radon is produced by the decay of radium in water or air. The hazard of radon gas is due its transporting from rocks into the environment soil to air by convection and diffusion through air or water circulation. The importance of each process depends on the geological nature of the formation. The exhalation rate of radon varies due to changes in differential air pressure, uranium and radium levels, working conditions and degree of ventilation. After exhalation, radon gas travels through ventilation streams and generates the solid decay products: Po-218, Bi-214 and Po-214 [10, 11].

The aim of this study is assessing of radioactive contamination in soil samples from Diyala Bridge area, Baghdad, using gamma-ray spectroscopy analysis and calculate radon gas inhalation dose.

2. Materials and methods

2.1. Collection and preparation of samples

This study was conducted in the Baghdad governorate in the area of Diyala Bridge. Soil samples were collected from twelve points taken from different locations in the area, as shown in Fig.1 and Table 1. One kilogram of soil samples was collected from each point, specifically from the topsoil at depths ranging from 0 to 5 cm. These soil samples were prepared for a series of tests in the laboratory. First, it was crushed and then dried in an oven at a temperature of 105 degrees Celsius for 8 hours. Then, the samples were sieved through a 300 µm mesh sieve. From the resulting homogenous soil samples, 1000 grams were carefully packed into a polyethylene beaker, weighed, sealed, and stored for a minimum of 4 weeks before further analysis. This waiting period allowed sufficient time for the isotopes of uranium-238 and thorium-232 to reach equilibrium with their respective radionuclide daughters.

Table 1: Numbers and codes of soil samples locations.

Number	Location	X coordinates	Y coordinates
1	Ibn-Alkatteeb Hospital	454555	3676350
2	Street Number21	454707	3676160
3	Jisr Diyala Police Station	454980	3677070
4	Alnafs Al-zakia School	456587	3675960
5	Near the gate of the Iraqi Atomic Energy Commission	455953	3675260
6	Mosque Asihab Al-kasa	457436	3676240
7	Musa Al-Kazem School	456909	3676750
8	Al-Basaer School	455279	3677070
9	Kaab bin Zuhair School	456202	3678060
10	Mosque Al-Rasul Al'aezam	456019	3676610
11	Al-Riyadh Model Hospital	456487	3677310
12	Khatam Al-Nabiiyn School	455656	3675910

Figure 1: The map of Diyala Bridge showing the sample location according to Arc GIS 10.8 program.

For the measurement of radioactivity, scintillation gamma spectrometry, extensively detailed by Killeen, was employed. This method is widely utilized for the determination of

uranium, thorium, and potassium in geological material samples. In the current study, the natural radioactivity present in the soil of the region was assessed using a gamma ray spectrometry setup utilizing a (3 × 3) inches NaI(T1) detector (ORTEC- model 905-4). The achieved regulation was typically <7.5% for the 662 keV gamma ray emitted by Cs-137. The calibration of gamma spectroscopy was done using standard isotopes such as ^{214}Am, ^{109}Cd, ^{137}Cs and ^{60}Co as present in Table 2.

In this study, specific energy photopeak were utilized for the quantitative determination of uranium, thorium, and potassium. The 1.76 MeV peak of Bismuth-214 was utilized for the quantitative determination of uranium, the 2.62 MeV peak of Thallium-208 was employed for the quantitative determination of thorium, and the 1.46 MeV peak corresponding to radioactive potassium was utilized for the quantitative determination of potassium. Each sample was counted for duration of 3600 seconds, and background counts were obtained under identical conditions which were subtracted from the gamma peaks.

Table 2: Radionuclides of Calibration Certificate

Radionuclide	Half-life(days)	Energy(keV)	Efficiency	Combined Standard Uncertainty%
^{214}Am	157800	59.53	0.106	1.1
^{109}Cd	462.6	88.34	0.628	1.5
^{137}Cs	11019	661.7	0.0709	1.2
^{60}Co	1925.4	1173.2 1332.5	0.0107	1.1

3. Calculations

3.1 Specific Activity

Equation (1) was used to find the specific activity(A) in the soil samples [12, 13]:

$$\boldsymbol{A\left(\frac{Bq}{kg}\right) = \frac{Neat\ Area}{t(s) \times I_\gamma \times \varepsilon \times M(kg)}} \quad (\mathbf{1})$$

where, Neat Area represents the net area of the photo-peak after subtracting the background radiation peak, t is the time of measurement (3600 s), Iγ is gamma- decay probability, ε is efficiency of NaI(Tl) detector for each gamma energy of the radionuclides found in soil, and M (kg) is the soil sample mass.

3.2 Calculation the Hazard Index by Radiation

3.2.1 Radium Equivalent Activity (Ra_{eq})

Radium Equivalent Activity (Ra_{eq}) is a measurement used to quantify the combined radioactivity of radium isotopes present in the sample. The value of Ra_{eq} is determined, based on the specific activity of the radioactive isotope radium-226 (Ra-226). This measurement helps to assess the potential radiation hazard, associated with materials containing radium and its decay products ^{226}Ra, ^{232}Th and ^{40}K, using Equation (2) [14]:

$$\boldsymbol{Ra_{eq}\left(\frac{Bq}{kg}\right) = A_{Ra} + 1.43A_{Th} + 0.077A_K} \quad (\mathbf{2})$$

where A_{Ra}, A_{Th}, A_K is the specific activity for ^{226}Ra (^{238}U), ^{232}Th and ^{40}K, respectively.

3.2.2 Gamma Rate of Dose Absorption (Dγ

Equation (3) was used to calculate the Gamma Rate of Dose Absorption (Dγ), [14, 15]:

$$\boldsymbol{DY(nGyh^{-1}) = 0.462A_{Ra} + 0.621A_{Th} + 0.0417A_K} \quad (\mathbf{3})$$

3.2.3 External Hazard Index (H_{ext})

The external hazard index (H_{ext}) is determined by Equation (4) [16]:

$$\boldsymbol{H_{ext} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}} \quad (4)$$

3.2.4 Internal Hazard Index (H_{int})

Internal Hazard Index (H_{int}) was calculated using Equation (5) [17]:

$$\boldsymbol{H_{int} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}} \quad (5)$$

3.2.5 Index of Gamma Representative (Iγ)

The Index of gamma representative (Iγ) can be determined using Equation (6) [15, 18]:

$$\boldsymbol{I_\gamma = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} = 1} \quad (6)$$

3.2.6 Annual Effective Dose Equivalent (AEDE)

The Annual Effective Dose Equivalent (AEDE) was determined by calculating AEDE values for indoor, outdoor, and total exposures using Equations (7), (8), and (9), respectively, using 8760 hr in year, 0.7 to convert the absorbed dose (Gy) to effective dose in air (Sv) and (0.8 and 0.2) the indoor and the outdoor fraction occupancy factors, and as outlined below [19, 20]:

$$\boldsymbol{AEDE_{indoor}\left(\frac{mSv}{y}\right) = D_Y\left(\frac{nGy}{hr}\right) \times 8760\left(\frac{hr}{y}\right) \times 0.7\left(\frac{Sv}{Gy}\right) \times 0.8 \times 10^{-6}} \quad (7)$$

$$\boldsymbol{AEDE_{outdoor}\left(\frac{mSv}{y}\right) = D_Y\left(\frac{nGy}{hr}\right) \times 8760\left(\frac{hr}{y}\right) \times 0.7\left(\frac{Sv}{Gy}\right) \times 0.2 \times 10^{-6}} \quad (8)$$

$$\boldsymbol{AEDE_{total}\left(\frac{mSv}{y}\right) = AEDE_{indoor}\left(\frac{mSv}{y}\right) + AEDE_{outdoor}\left(\frac{mSv}{y}\right)} \quad (9)$$

3.2.7 Excess Life Time Cancer Rate (ELCR)

The Excess Lifetime Cancer Rate (ELCR) can be expressed using the following mathematical equation:

$$\boldsymbol{ELCR = AEDE\ total \times D_L \times R_F} \quad (10)$$

where D_L represents the average period of lifetime, typically estimated to be around 70 years, and R_F is the conversion factor. The International Commission on Radiological Protection (ICRP) employs a standard R_F value of 0.05 Sv^{-1} for the public [21, 22].

3.3 Calculation the concentration of ^{222}Rn

The radioactive-concentration of ^{222}Rn in soil samples was estimated by measuring the radioactive concentration for ^{222}Rn present in air surrounding soil samples, as described in UNSCEAR, which involves several steps [23]:

First, evaluating the radioactive concentration for ^{222}Rn within the soil samples, using Equation (11):

$$\boldsymbol{Gs(n) = F_r \times \rho \times C_{Ra}(n)} \quad (11)$$

Where Gs(n) is the specific activity of radon gas inside the soil sample (n) (Bq/m^3), F_r represents the emission constant of ^{222}Rn for soil and equal (0.1), ρ is the soil density and equals (1800 kg/m^3), $C_{Ra}(n)$ is the specific activity for ^{226}Ra soil sample (n) (Bq/kg).

Now specific activity of ^{222}Rn in air can be calculated by Equation (12):

$$\boldsymbol{Ca(n) = Gs(n)\ (d_{soil} / D_{air})^{1/2}} \quad (12)$$

Where Ca(n) is the specific activity of ^{222}Rn the air for sample (n) (Bq/m^3), d_{soil} is the diffusion rate constant of ^{222}Rn soil (0.5×10^{-4} m^2/sec), D_{air} is the diffusion rate constant of ^{222}Rn air (5 m^2/sec), D is the fixing dosages rates from radon gas inhalation, and vegetable consuming determined using the equation [24].

After that, dose rate resulting inhalation of radon gas consuming (Hp) expressed for (Sv/y) can calculated using following equation:

$$\mathbf{Hp = Ca(n) \times Ip \times DCF} \quad \mathbf{(13)}$$

Where Ip is the amount of consumption in outdoor air (600 m^3/year) [24], DCF is the dose transformation coefficient for ^{222}Rn equal (1.3×10^{-9} Sv/Bq) [23].

4. Results and Discussion

Table 3 presents the specific activity values (Bq/kg) in soil samples for the radionuclides: Ra-226 (186 keV), Bi-214 (609 keV), Cs-137 (662 keV), Ac-228 (969 keV) and K-40 (1460keV).

Table 3: Specific Activity (Bq/kg) values in soil samples for Radionuclides: Ra-226 (186 keV), Bi-214 (609 keV), Cs-137 (662 keV), Ac-228 (969 keV) and K-40 (1460keV)

No. of sample	Specific Activity for Radionuclides (Bq/kg)				
	Ra-226 (U-238) 186 keV	Bi-214 (U-238) 609 keV	Cs-137 662 keV	Ac-228 (Th-232) 969 keV	K-40 1460keV
1	47.3	0	0	0	427
2	21.0	19	50	0	323
3	76.1	0	39	20.5	252
4	0	0	0	71.3	158
5	263.4	0	102	0	108
6	89.2	20	38	15.7	200
7	65.3	12	19	11.4	409
8	22.3	8	18.3	12.9	302
9	15.9	16.9	68	12	141
10	26.3	0	42.8	16.7	224.8
11	80.3	32.7	72	24	377.7
12	112.4	15.9	45.7	19.7	223.8
Mean	68.3	10.4	41.2	17.0	328.9
Permissible Limit UNSCEAR (2010) [26]	-----	-----	-----	-----	412

From Table 3, it is found that, the lowest specific activity of K-40 was (108) Bq/kg in sample (5), while the maximum specific activity was (958) Bq/kg in sample (4), the mean value of K-40 for the total number of samples was (328.9) Bq/kg. The minimum specific activity in Ra-226 was (0) Bq/kg in sample (4), whilst the highest specific activity of (263.4) Bq/kg was for sample (5). The average for the total number of samples was (68.3) Bq/kg. The lowest specify activities of Th-232 (Ac-228) was (0) Bq/kg for samples (1,2, and 5), whilst the highest was specific activity of (71.3) Bq/kg in sample (4), the mean specify activity of all samples was (17.0) Bq/kg.

The calculated values of hazard index in soil samples Ra_{eq} (Bq/kg), $A_D\ (nGyh^{-1})$ H_{ex}, H_{in} , $I\gamma$, $AEDE_{in} \times 10^{-6}\left(\frac{Sv}{Gy}\right)$, $AEDE_{out} \times 10^{-6}\left(\frac{Sv}{Gy}\right)$, $AEDE_{total} \times 10^{-6}\left(\frac{Sv}{Gy}\right)$ and ELCR $10^{-6}(\frac{Sv}{Gy})$ are shown in Table 4.

Table 4: The values of hazard index in soil samples Ra_{eq} (Bq/kg), $A_D\ (nGyh^{-1})$ H_{ex}, H_{in} , $I\gamma$, $AEDE_{in} \times 10^{-3}\left(\frac{Sv}{Gy}\right)$, $AEDE_{out} \times 10^{-3}\left(\frac{Sv}{Gy}\right)$, $AEDE_{total} \times 10^{-3}\left(\frac{Sv}{Gy}\right)$ and ELCR $10^{-3}(\frac{Sv}{Gy})$

No. of sample	Ra_{eq} Bq/kg	A_D $(nGyh^{-1}$	H_{ex}	H_{in}	I_γ	$AEDE_{in}$ $10^{-3}(\frac{Sv}{Gy})$	$AEDE_{out}$ $10^{-3}(\frac{Sv}{Gy})$	$AEDE_{total}$ $10^{-3}(\frac{Sv}{Gy})$	ELCR 10^{-3}
1	80.179	41.964	0.217	0.344	0.600	0.206	0.051	0.257	0.901
2	45.871	24.915	0.124	0.181	0.355	0.122	0.031	0.153	0.535
3	124.819	59.758	0.337	0.543	0.880	0.293	0.073	0.366	1.283
4	175.725	89.399	0.474	0.474	1.352	0.439	0.110	0.548	1.919
5	271.716	126.778	0.734	1.446	1.828	0.622	0.155	0.777	2.721
6	127.051	60.380	0.343	0.584	0.885	0.296	0.074	0.370	1.296
7	113.095	56.512	0.306	0.482	0.822	0.277	0.069	0.347	1.213
8	64.001	32.538	0.173	0.233	0.479	0.160	0.040	0.199	0.698
9	43.917	21.439	0.119	0.162	0.320	0.105	0.026	0.131	0.460
10	67.491	33.109	0.182	0.253	0.492	0.162	0.041	0.203	0.711
11	143.703	69.792	0.388	0.605	1.027	0.342	0.086	0.428	1.498
12	157.804	74.703	0.426	0.730	1.096	0.366	0.092	0.458	1.603
Mean	117.948	57.607	0.319	0.503	0.845	0.283	0.071	0.353	1.236
Permissible Limit UNSCEAR (2000) [25]	370	--------	≤1	≤1	1	--------	--------	0.083×10^{-3}	0.29×10^{-3}

Table 4 shows that the lowest calculated radium equivalent (Ra_{eq}) using Equation (2), was equal to (43.917) Bq/kg in sample 9, the highest value was 271 in sample (5) with an average rate of (117.948) Bq/kg. Using Equation (3), the absorption dose rate of Gamma radiation (D_γ) was determined. The highest calculated dose rate was 126.778 nGy/h, while the lowest was 21.439 nGy/h, with an average rate of 57.607 nGy/h.
(H_{ex}) was calculated using Equation (4), the highest H_{ex} value was 0.734, below the allowable limit of 1.00. The lowest H_{ex} value was 0.119, with an average of 0.319.
(H_{in}) was obtained using Equation (5), the highest H_{in} value was 1.446, while the lowest was 0.162, with an average of 0.503.

(Iγ) was determined using Equation (6). The highest I_γ value was 1.828, and the lowest was 0.320, with an average of 0.845.
(AEDE) was calculated using Equation (7); the highest annual internal dose equivalent was 0.622 mSv/y, whilst the lowest was 0.105 mSv/y, mean of 0.283 mSv/y.
($AEDE_{out}$) was obtained using Equation (8), the highest outdoor equivalent dose was 0.155 mSv/year, the lowest value was 0.026 mSv/year, average 0.071 mSv/year, it was less than the allowed dose (1 mSv/year)[14]. ELCR$\times 10^{-3}$ values ranged from 0.460 to 2.721, with a mean value of 1.236.

The ELCR and $AEDE_{total}$ increased as the radium equivalent and absorbed dose increased. However, the values of Ra_{eq} were lower than the recommended values, but the values of ELCR were higher than the allowed value (0.29×10^{-3}).
Table 4 presents the concentration of ^{226}Ra in soil, concentration of radon emitted from ^{226}Ra in soil Gs(n) (Bq/m^3) , the concentration of radon liberated in air from ^{226}Ra (Ca(n) (Bq/m^3)), and dose average (Hp) resulting from inhalation of gas radon consumption (Sv/y) in soil samples.

Table 5: Concentration of 226Ra in soil, the concentration of Radon emission from 226Ra in soil Gs(n) (Bq/m3) , the concentration of Radon liberated in air from 226Ra (Ca(n) (Bq/m3)) and dose average (Hp) resulting inhalation of gas radon consumption (Sv/y) in soil Samples.

No. of sample	Ra-226 186 keV	Gs(n) (Bq/m^3) In soil	Ca(n) (Bq/m^3) in air	Hp (Sv/y) $\times 10^{-9}$
1	80.179	8514	0.043	33.205
2	45.871	3780	0.019	14.742
3	124.819	13698	0.068	53.422
4	175.725	BDL	BDL	BDL
5	271.716	47412	0.237	184.907
6	127.051	16056	0.080	62.618
7	113.095	11754	0.059	45.841
8	64.001	4014	0.020	15.655
9	43.917	2862	0.014	11.162
10	67.491	4734	0.024	18.463
11	143.703	14454	0.072	56.371
12	157.804	20232	0.101	78.905
Mean	**117.948**	**12292.5**	**0.061**	**47.941**

It was found that contamination of the soil with one or all of the radioactive chains leads to contamination of the soil and the air surrounding it with radon gas, and that the highest absorbed dose as a result of inhaling air containing radon is 184.907×10^{-9} (Sv/y) in sample 5, and that the lowest absorbed dose is in the sample 9 was 11.162×10^{-9} (Sv/y).
Figure 2 shows that the higher the concentrations of radon gas in the air, the greater the life time cancer rate. As the highest value was in sample 5 and the lowest value was in sample 9.

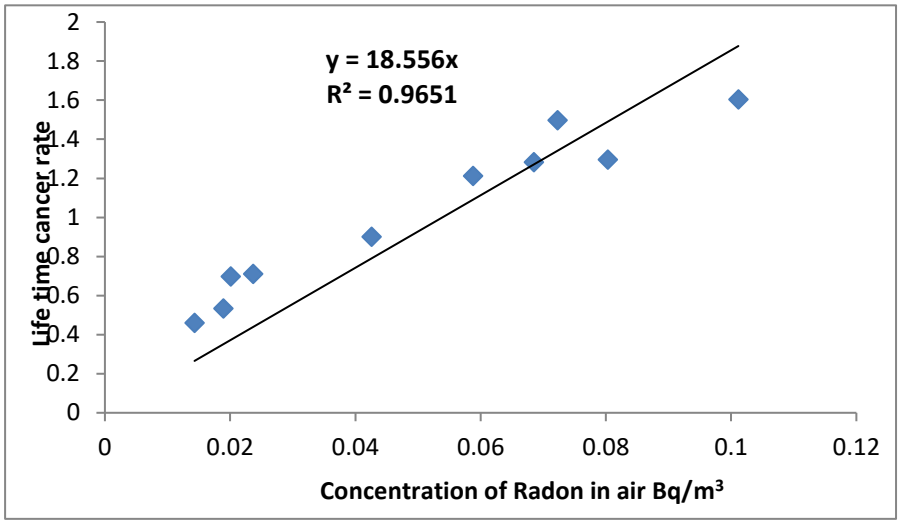

Figure 2: Life time cancer rate and concentration of Radon in air in soil samples.

5. Conclusions

1- This investigation reveals that soil samples collected from areas around Diyala Bridge / Baghdad exhibited varying levels of the radioactive isotope Ra-226, with notable exceptions observed in sample 4.

2- Analysis of γ-absorbed dose rates and natural radioactivity stemming from soil samples and air indicates that the majority of the studied samples pose a radiological hazard, with the exception of samples 2 and 9.

3- Despite the presence of radioactive isotopes, the soil is generally deemed safe for use in construction and agricultural activities, posing minimal radiation risk to the populace. However, caution is warranted regarding samples 2 and 9.

4- Specific activity levels for all radionuclides fall below permissible limits, with the exception of a region near the Iraqi Atomic Energy facility in Al-Twetha city.

5- Soil contamination by radioactive elements results in the emission of radon gas, impacting both the soil and surrounding air. The average absorbed dose from inhaling radon-containing air is calculated to be 47.941×10^{-9}(Sv/y).

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