Soil Measurements Using High Purity Germanium Gamma Spectroscopy for Areas Surrounding Al-Tuwaitha Nuclear Site

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Abstract

This study introduces the calculations of the radiation background, by characterizing the radionuclides and calculating their specific effectiveness in the environment of the city of Baghdad (soil and plants) using the gamma-ray spectroscopy technique. 20 soil samples at a depth of 20 cm were collected for 5 locations on the Rusafa side, where the site coordinates were fixed using the G.P.S device. A modern global gamma-ray spectroscopy system (DSA 2000) was used, with high-purity germanium (HPGe) detector with an efficiency of 50% and an analysis capacity of 2.2 keV with respect to the energy of 1332 keV for the ^{60}Co source. The results of the specific activity rates of the radionuclides (^{214}Bi or ^{214}Pb), (^{228}Ac or ^{208}Tl), (^{40}K), and (^{137}Cs) in soil were: (50.735 Bq/kg or 0.525 Bq/kg) which is close to the value of the previously published results, as the highest value was recorded in Bangladesh (88.1 Bq/kg or 4.8 Bq/kg). The results of the qualitative effectiveness of the soil of the sites showed normal levels within the radiation background of Baghdad's soil. There was a significant convergence of the study's findings with other studies.

1. Introduction

Natural radioactivity, known as radiation background, is crucial for the population's exposure to radiation. Many states in the world measure the rate of exposure caused by natural radiation for various purposes, including the selection of nuclear installations locations, contingency plans to monitor any case of increased radioactivity, and identifying its source and cause to ensure the security of the state and citizens.

Measurements of the exposure rate began nearly 20 years ago as part of the country's radioactivity environmental control program, and the health impacts of such environmental radioactive contamination can be accessed through the estimation of radiation traction resulting from different population exposures and the use of internationally approved hazardous functions.

To assess the biological impact on human beings, it is necessary to calculate radiation doses originating from radioactive materials, their pathways, and mitigating factors in the environment, through natural phenomena such as wind, rainfall rate, soil nature, topography, evaporation rate, and temperatures. Knowledge of how soil and plant factors and environmental conditions affect radionuclide transmission is essential to reduce their risk to

the environment in general and in particular to human's radionuclides from soil to plant can be explained by the modulus of transport, which represents the ratio between the concentrations of radionuclides in the plant to its concentration in the soil. Different radioactive elements of the environment behave in a manner similar to those of non-radioactive elements, connecting the daily food chain depending on the chemical and physical properties of these elements, then entering the human body causing biological damage (depends on the radiation quality factor, and the effective equivalent dose of the tissue) to the tissue and organs targeted depending on the "equivalent dose "of those organs and their ability to store and handle those elements [1].

Potassium ^{40}K, which is a natural radioisotope, is found to contribute the largest part of people's internal radiation occupied dose. In the event of a nuclear explosions or nuclear accidents, the radioactive sources spreading into the atmosphere and their sediments above the earth's surface are liberated by the so-called "fallen". The most important fission products, such as Strontium ^{90}Sr with a half-life of 28.8 years, and Cesium ^{137}Cs with a half-life of 30 years, possess relatively long half-lives and are similar in their chemical composition to the natural composition of the body's core elements. Strontium ^{90}Sr is similar to Calcium that represents an essential element of the bone composition, and belongs to a group (1) of the periodic table, while Cesium ^{137}Cs is similar to Potassium that belongs to a group (2) of the periodic table, and is considered as a cornerstone of the structure of muscles cells. Accordingly, the pollution will be relatively greater.

There are numerous studies in which this detector has been used to study environmental contamination with radioisotopes since the 1950s and 1960s, when the first nuclear tests began, followed by nuclear accidents and the important Chernobyl accident in 1986 in Ukraine, as a broad area of researches around the world.

There are numerous studies related to the environmental pollution of southern Iraq. (1998 ornaments) Studied models of air, soil, and plant in selected areas of southern Iraq and studied radioisotope transmission factors from soil to plant. The total annual dose of the whole body at 441.6 μSvy-1 is calculated. The researcher stated that the dose from inhalation is 99% of the dose of the whole body. The comparison between the dose received before and after 1991 indicates that the amount of the dose in 1991. It was much higher than the dose before that date [2].

A conducted a study on the impact of radiological weapons in the areas of warfare operations in southern Iraq using the high-purity germanium Spectroscopy on a range of environmental mode (animal and plant tissue) and tested the presence of high concentrations of Radium ^{226}Ra and Thorium ^{234}Th in soil and weed. In other sample of plants, the concentration of 214Bi Bismuth and lead 214Pb is three times greater than in Measurements taken from areas far from the Attacked zone.

The Uranium concentration studies in groundwater and surface in the Military operation area. The tests established a rise in Radium concentration in sediment samples collected from water channels in Jabal Al-Senam located in south of Rumaila near Iraq-Kuweit border, areas close to the presence of bombarded armors. The concentration rate was 94 Bq kg^{-1}, twice the normal concentration of that area (field measurements).

In another study, it was used   reverse analysis to find radionuclide concentrations in areas in southern Iraq [3]. including Safwan, Zubair, Rumaila North and South, and Jabal Sanam, in which depleted Uranium weapons were used during the 1991 Gulf War. Areas of soil contaminated from 1991 to 1996 were found at 1718 km^2 as well as the amount of water that was transferred during its run-off as a torrent for the transport of radioactive contaminants to nearby water channels at 1886,103 m3. River floor deposits contaminated with radioisotopes, especially radium ^{226}Ra amounted to 14 km^2. The researchers attributed the increase in external exposure as a result to radioactivity and its increased normal emission of radon ^{222}Rn from the destroyed armors and the contaminated areas. Researchers studied and calculated the concentrations of the aforementioned sources at nuclear accident sites as the projections of the Chernobyl accident reached northern Iraq and the first Gulf War in which depleted Uranium Weapons were used.

In this study, the area around the Tuwaitha will be taken, the areas around nuclear site because of the looting and tampering of radioactive materials that existed before 2003 so that we can see the impact of that chaos on these areas.

2. Material and Methods

After completing the collection of maps of the Tuwaitha site in shown in Figure (1), nuclear sites such as reactors, isotope production sites, and the treatment of radioactive waste are surrounded by soil embankment of 30-35 m height in order to secure control of these sites, thereby making the increase in the radiation background of the areas around the site either by evaporation or by unsafe transport or by tampering and chaos, and thus excluding the wind reaction. The process of collecting samples was:

(A) Collection of Soil Samples

After the data gathering form fields have been designated with numbers and symbols, the models were collected according to the IAEA's recommendations. At least ten samples were taken per zone, for different areas depending on the surroundings of the site and for four pillars per site, and at a depth of about 20 cm, and Samples were placed in plastic bags with codes for the area and numbers for the Sample location coordinate (GPS).

Figure (1). Map of Al Tuwaitha nuclear site facilities (2020) (by GIS Programme).

(B) Collecting Plant Samples

The areas are nominated to collect samples from the surrounding areas of the site as the seasonal plants, and permanent plants to study, the true concentrations of radioactive elements in the stems and leaves, where the contamination of vegetation by radioactive elements is a long-term threat to humanity, Therefore, plant models have been taken from the soil model sites themselves. Plants were cut at levels close to the ground surface of approximately 20 mm, representing the height of weeds normally consumed by livestock. After completely dehydrated, then removed the impurities, mechanically grinded, smoothed, and sifted with a 2 mm sieve, and they are ready for laboratory inspection.

(C) Model Analysis
High Purity Germanium Detector

To study the samples content of both Cesium ^{137}Cs and potassium ^{40}K, the high purity germanium detector was used in the analysis of the model containing radioactive elements. This requires important steps, the most important of which is to calibrate the energy and then set the peaks for the elements within the region of interest ROI and thus measure the area under the peaks. With a view to the effectiveness of the radioactive component, HPGe system was used for the measurement. A schematic diagram and a photo of the instrument are illustrated in Figures (2 & 3).

The spectrometry analyzer used in this study is an advanced type device CANBERRA used to analyze and study spectrometry, and relies on three micro-computers that control its memory and dashboard, only to enter and extract information between the device and the other used devices.
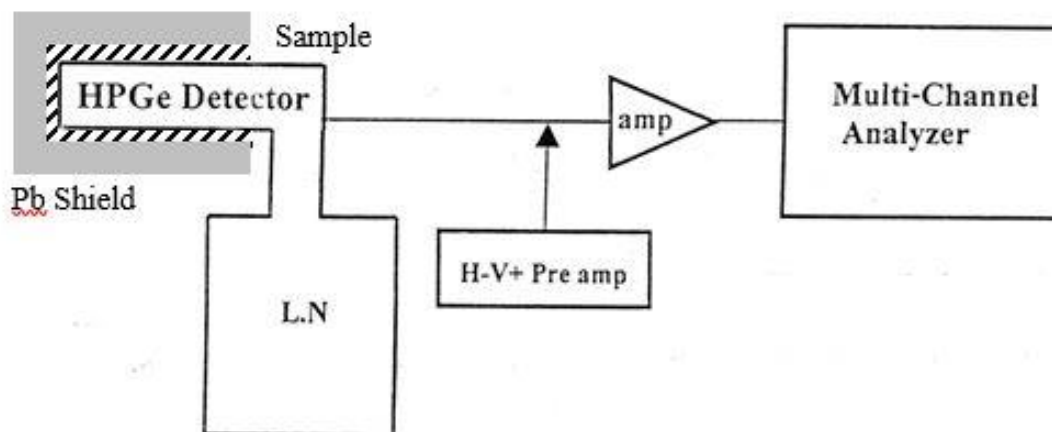

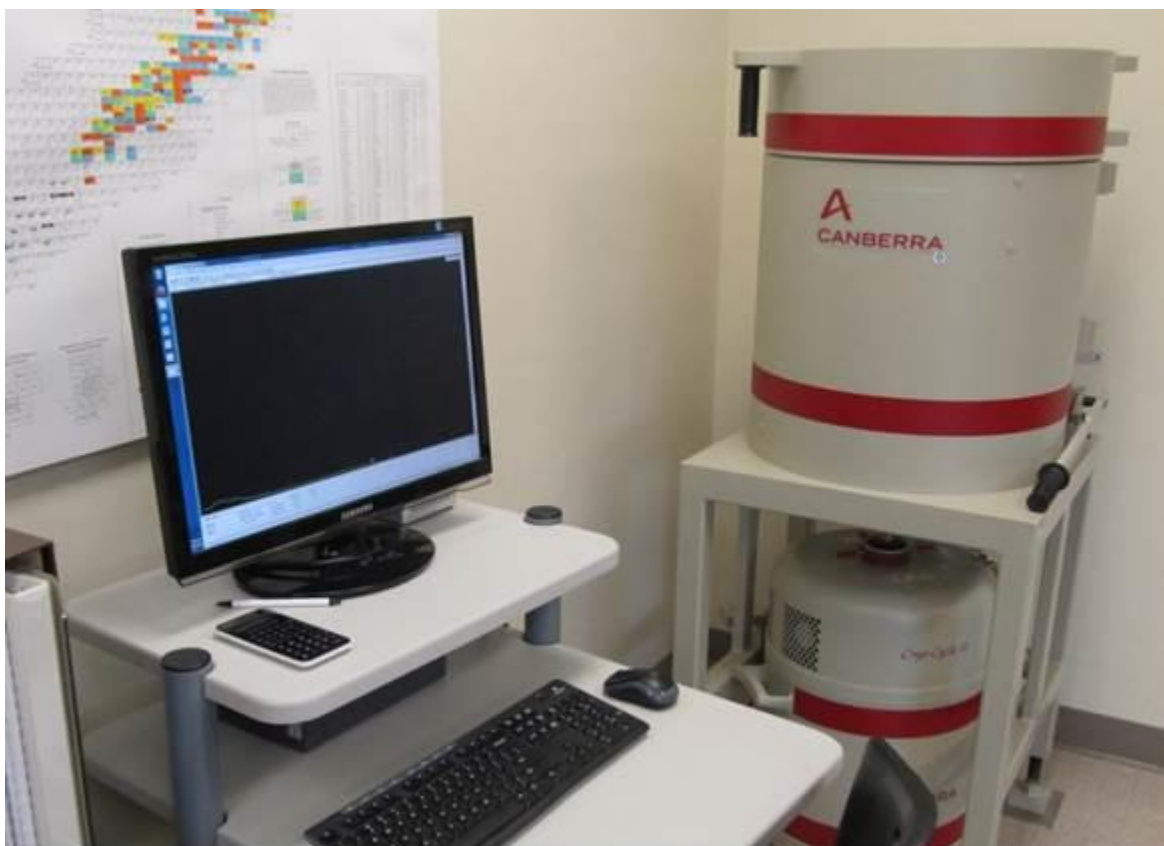**Figure (2).** The germanium spectroscopy system [4].

Figure (3). A photograph of HPGe system [4].

For spectrum analysis, the following steps must be followed:

Power Calibration
In the calibration process, a collection of radioactive calibration sources with well-known activities and gamma energy were used. These sources are "provided by" the International Atomic Energy Agency (IAEA). The calibration is done directly by the analyst using the "Least-square fit" method.

Table (1). The energy and intensity of the calibration sources.

Radioactive elements	Energy keV	Relative intensity %
Cs – 137	661.645	84.6
Co – 60	1173.238	99.87
Co – 60	1332.501	99.98
Ra – 226	186.200	8.07

Measuring Energy Separation Capacity
For better and optimal measurements of the detector's energy separation, its energy separation capability was measured as a function of the bias voltage difference, the magnifier formation constant, and the counting rate. The Cobalt 60Co isotope was placed 25 cm away from the detector surface which was used to measure the detectable susceptibility to the peak energy separation 1332 KeV as a function of its biased voltage difference, where the used voltage difference was 3200V throughout the study.

High Purity Germanium Detector Efficiency
Intrinsic efficiency is defined as the ratio between the numbers of pulses recorded to the radiation falling on the detector of the unit of time. Efficiency varies from one detector to another depending on two main factors: the type and dimension of the detector material and the type and energy of the radiation falling. The detector's efficiency increases with the increase of the probability of the reaction between the fallen radiation and the detector material and with the increase of the dimensions of the detector.

3. Results and Discussion
The measurement system for modern GENE 2000 (Software for samples analysis) has been used globally, and the duration of measurement has been selected according to the working conditions such as power outages. Table (2) shows the change of the detection limit of the selected energies over time for a random sample. (KL12), where it was first found that radionuclides were characterized by radionuclides (D) It does not change after the period (10000 sec), and secondly the detection limit does not change much is less than 15% for post-term reading (10000 sec). Figure (4) shows the change of detection limit as a function of time for different energies.

Table (2). The detection limit of selected energies changes over time for the sample KL12.

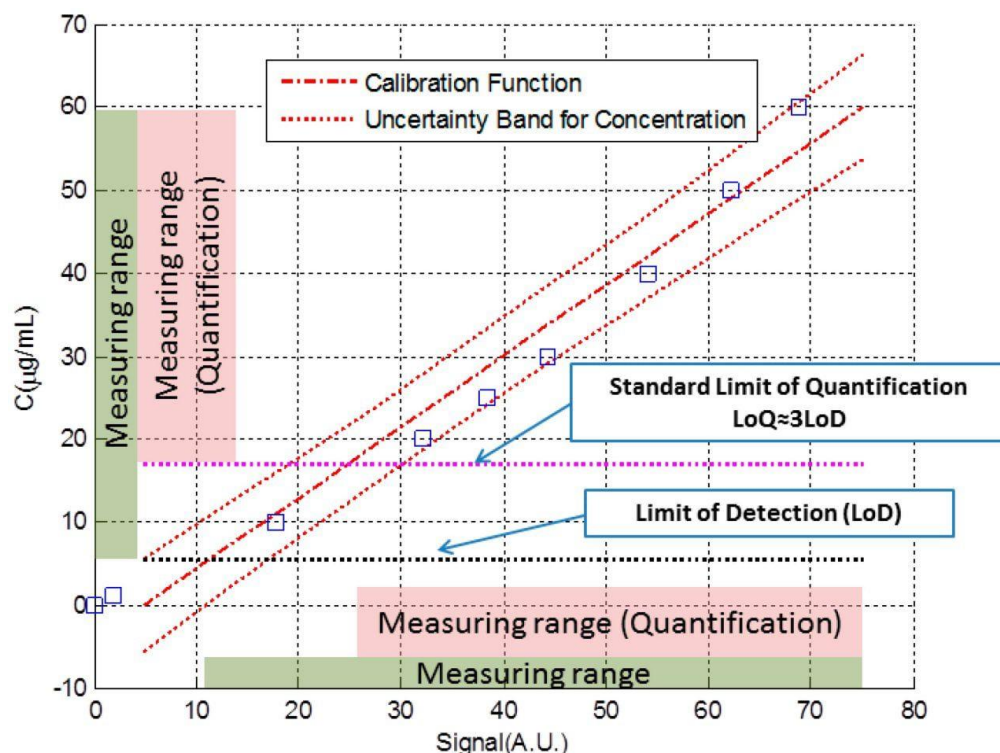| Time (sec)	Detection Limit (Bq/Kg)			
	K-40 1460 MeV	Ac-228 911 MeV	Bi-214 609 MeV	Pb-214 352 MeV
3600	17.49	10.31	5.52	6.35
7200	12.55	6.30	3.82	4.55
10000	11.04	6.28	3.60	4.16
14400	9.82	4.88	2.78	3.58
18000	8.74	4.22	2.70	3.27
20000	8.30	4.55	2.55	2.89

Figure (4). The change of detection limit over time for different energies. (GENE 2000) [6].

The qualitative effectiveness levels of Bismouth-214 or Lead-214 in topsoil models at their highest value (62.524±2.217) Bq/kg in the west of Tuwaitha area and at their lowest value (41.235±1.958) Bq/kg in the Riyadh area. The overall rate of quality effectiveness for Uranium at surface soil on the sidewalk side is (50735±0.525) Bq/kg. The qualitative effectiveness levels of Actinium-228 or Thallium-208 in topsoil showed the highest value (59.666±3.737) Bq/kg in Al-Wardia area, and the lowest value (39.029±3.120) Bq/kg in northern Tuwaitha site. The overall rate of quality effectiveness of Thorium in surface soil on the pavement side is (52.667 ± 0.838) Bq/kg. The qualitative effectiveness levels of Cesium-137 showed the highest value (24.085± 1.123) (Bq/kg) in Al-Wardia area, and the lowest value (2.332±0.633) Bq/kg in northern Tuwaitha site, the overall rate of quality effectiveness of this Isotope was (6.305±0.188) Bq/kg. The specific activity levels of cesium-137 nuclide in surface soil samples, and the value (24.085 ± 1.123) Bq/kg in the Zaafaraniya area, and the lowest value was (2.332 ± 0.633) Bq/kg in the Rashid camp area, and the overall rate of specific effectiveness for this nuclide was (6.305± 0.188) Bq/kg.

Table (3). Qualitative effectiveness levels of different nuclei in surface soil models on the sidewalk side.

Qualitative effective Bq/kg			Model Number	location
Cs-137	**Ac-228 or Tl-208****	**Pb-214* or Bi-214**		
5.170 ± 0.912	50.579 ± 3.735	62.524 ± 2.217	W.T.	Western Tuwaitha
24.085 ± 1.123	59.666 ± 3.737	56.937 ± 2.141	A.W.	Al-Wardia
4.464 ± 0.482	46.445 ± 3.025	46.175 ± 2.442	J.A.	Jaber Al-Ansari neighbor
4.325 ± 0.683	51.487 ± 3.474	51.910 ± 1.899	A.S.	Al-Salam neighbor
9.812 ± 0.887	53.327 ± 3.647	41.235 ± 1.958	A.R.	Al- Riyadh
2.332 ± 0.633	39.029 ± 3.120	47.391 ± 1.903	N.T.	Northern Tuwaitha

3. Conclusions

Results of quantitative effectiveness measurements of radionuclides were obtained in environmental samples of the areas around the Tuwaitha site, which included soil and plant. This study has been characterized by the inclusiveness of most elements of the environment such as soil and plant, which require considerable scientific and material efforts to measure the natural radiation effectiveness. Radiological background measurements of the environment of the areas around the Tuwaitha nuclear site will be the starting point for future works if we know that most Iraqi studies have not been characterized by such a comprehensive method. Our used system is considered a modern system with high specifications and accuracy of measurements and it was first used in Iraq. For Actinium-228 and Thallium-208 equivalent to Thorium-232, the readout rate measured values for different models are indicated as follows:

soils surrounding the Tuwaitha site (50.323±0.608) Bq/kg. It approaches the average value of as the highest value (70.30±16.63) Bq/kg in Malaysia's soil and the lowest value (14.8) Bq/kg in Russia. While in most studies Cesium-137 levels indicate that the qualitative effectiveness of this nucleus as an industrial product and most of the published qualitative effectiveness levels is an approach to the results obtained in our study where the sediment (7.619±0.255) Bq/kg and the soil of the surrounding areas (9.733±0.156) Bq/kg conclude that the radiation background of the surrounding areas around the Tuwaitha site is within normal range.

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