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Natural Radioactivity and Associated Dose Rates in Soil Samples in the Destroyed Fuel Fabrication Facility, Iraq

Abdulla Ahmad Rasheed[1], Nada Farhan Kadhum[1], Nadhim Khaleel Ibrahim[2,*]

[1]Department of Physics, College of Science, AL-Mustansiriya University, Iraq
[2]Ministry of Science and Technology, Iraq
*Corresponding author: nawi1980@yahoo.com

Abstract The activity concentrations of naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K in soil samples collected from Fuel Fabrication Facility in Al-Tuwaitha nuclear site were measured by using a high resolution gamma spectrometry system via High Purity Germanium (HPGe) Detector with a relative efficiency of (>40%) and resolution (<1.8keV) at energy of (1.33MeV) for ^{60}Co. The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil samples were found to be 15.78±1.16, 14.09±1.33 and 306.42±18.1 Bqkg^{-1}, respectively. The results obtained for the corresponding radionuclides are lower than the worldwide average values of 35, 30, and 400 Bqkg^{-1}, respectively. The average absorbed dose rate in air (Dγ), the average radium equivalent activity (Ra_{eq}) and the average external hazard index (H_{ex}) were determined as 28.90±2.12 nGyh^{-1}, 59.52±4.45 Bqkg^{-1} and 0.16±0.012, respectively, which are below the permissible limit.

Keywords: *activity concentration, gamma spectrometry, radium equivalent activity, hazard indices*

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1. Introduction

Exposure to ionizing radiation from natural sources is a continuous and unavoidable feature of life. Human beings are exposed to natural background radiation every day from the ground, building materials, air, food, outer space, and even elements in their own bodies. Gamma radiation emitted from primordial radionuclides and their progenies are ones of the main external sources of radiation exposure to the humans [1].

There are many activities increasing the radioactive exposure from natural sources of radiation, such as, mining and use of ores containing naturally radioactive materials, the radioactive residues resulting from nuclear weapons testing, the use of radioactive materials in industry and medicine, and operation and decommissioning of the nuclear power plants and other nuclear facilities [2].

The natural radioactivity in the environment is the main source of radiation exposure for human body. Natural radionuclide in soil contributes a significant amount of background radiation exposure to the population through inhalation and ingestion. The main contributors of radionuclides are ^{226}Ra, ^{232}Th and ^{40}K. Since these radionuclides in soils are not uniformly distributed and vary from region to another [3].

2. Materials and Methods

2.1. Study Area

The Fuel Fabrication Facility (FFF) is one of many facilities in Al-Tuwaitha site. Al-Tuwaitha Nuclear Research Center (ATNRC) is the principal nuclear site in Iraq and covers an area about 1.3 km^2 and is located approximately 1 km east of the Tigris River 18 km south of Baghdad. This site is fortified by large earthen berms around the facilities which cover over one km^2 [4,5]. Figure 1 shows Al-Tuwaitha site map location and the building layouts within Al Tuwaitha and the associated sector names. Al-Tuwaitha site was heavily bombed during the Gulf War of 1991, and most of the facilities were extensively damaged [6].

2.2. Samples Collection

Ten (10) soil samples were collected from different location inside the Fuel Fabrication Facility at 5 to 10 cm depth from the top surface soil layer to make approximately 1 kg weight per sample, each soil sample is filled into secure polyethylene bag to prevent cross contamination and sent to the laboratory.

2.3. Samples Preparation

Sample preparation is carried out by placing each soil sample in an oven for drying at a temperature of 80°C for 2h, thus ensuring complete removal of any residual moisture. The dried samples were pulverized into a fine powder. To obtain uniform particle sizes, a 500 µm mesh is then used to sieve the samples which are transferred to 500 ml labeled Marinelli beakers and weighed, sealed and labeled. The samples are stored for at least one month in order to maintain secular equilibrium between ^{238}U, ^{226}Ra, and ^{232}Th and their short-lived progeny. The samples were each counted on a High Purity Germanium Detector for one hour (3600s).

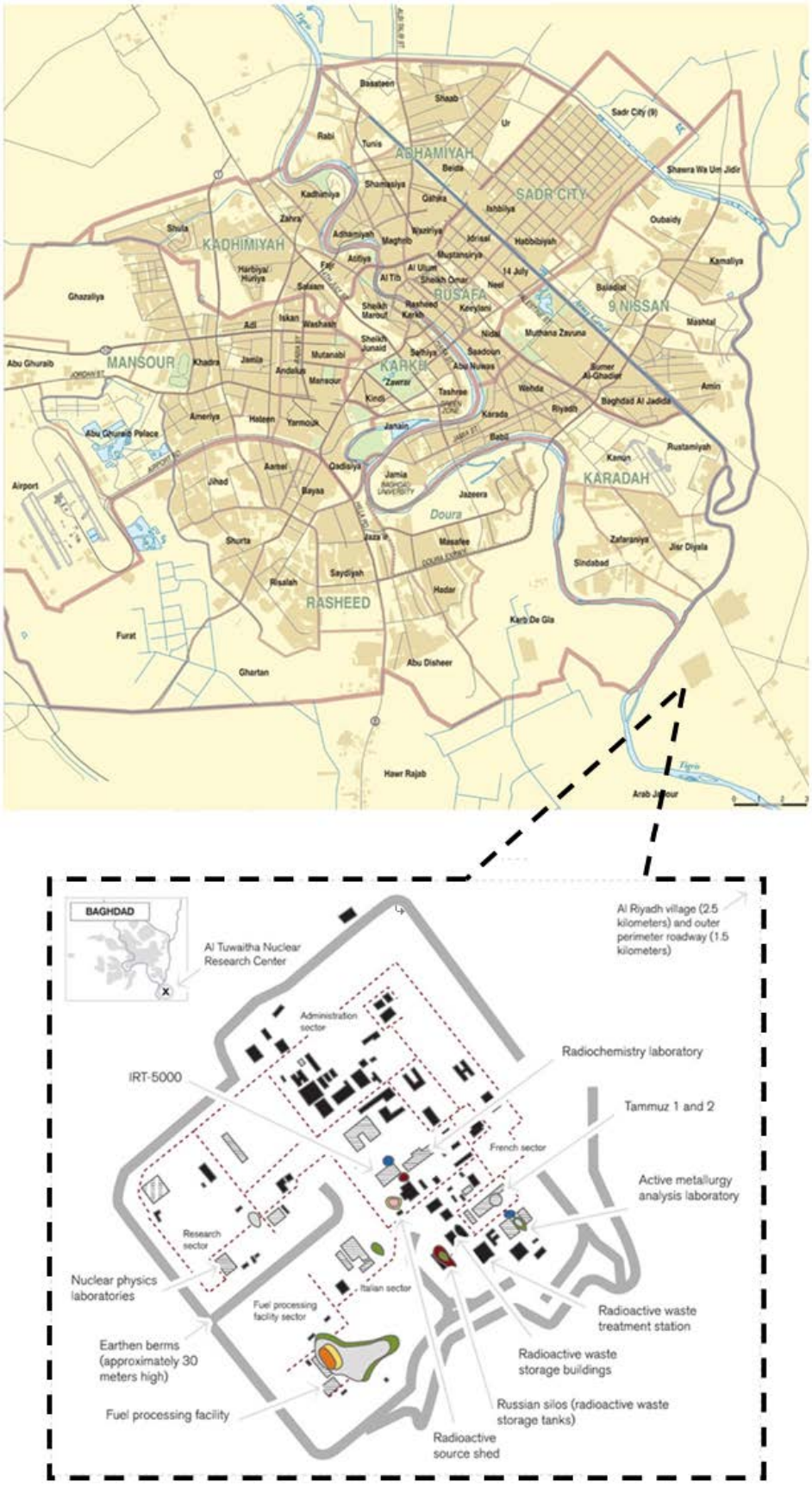

Figure 1. Al-Tuwaitha site map location, Baghdad, Iraq

2.4. Gamma Spectroscopy System

In this work, gamma spectroscopy technique has been used for measuring the specific activity concentration of radionuclides. For certain radionuclides that cannot be effectively measured directly in the field, soil samples of the area under investigation should be collected and then analyzed to determine the radionuclide concentration with a laboratory-based procedure. All processes in this work have been performed in Central Laboratory Directorate

(CLD) at Ministry of Science and Technology (MoST) of Iraq.

2.4.1. High Purity Germanium (HPGe) Detector

A pure germanium closed end-coaxial P-type detector manufactured by CANBERRA Company (model GC4018) with a relative efficiency of (>40%) and resolution (<1.8keV) at energy of (1.33MeV) for 60Co. It contains a HPGe crystal has a diameter of 62mm, a length of 60mm and a distance from window 4.67mm. It is surrounded with lead shielding 10cm thickness to reduce the background radiation from various natural radiation sources and to isolate from other radiation sources used in nearby surroundings.

2.4.2. Calibration of the Gamma Spectrometry System

In order to obtain any meaningful results from a gamma spectrometry system, such as isotope identification, qualitative and quantitative analysis, the system must be calibrated both in terms of energy as well as efficiency. Normally calibration is done with standard sources and reference material of known activity.

The energy calibration related channel number of the spectrometer to the energy of the standard reference material. The calibration was performed by matching principal gamma rays in the spectrum of the standard to the channel numbers.

A standard source of multi-gamma energy has been used to calibrate the efficiency of the detector. Figure 2 shows the efficiency calibration curve for the (HPGe) detector using mixed standard source in the 500 ml Marinelli beaker by acquiring spectrum for one hour (3600s) with different energy ranges from 59.5 keV for ^{241}Am to 1836.06 keV for ^{88}Y.

The net count rate was determined at photopeaks for all energies used for determining the efficiency. The efficiency is related to the count rate of standard [7] as:

$$\varepsilon\left(E_\gamma\right)=\frac{N}{AI_\gamma\left(E_\gamma\right)t}\times 100\% \quad (1)$$

Where:

$\varepsilon(E_\gamma)$ is the detection efficiency at energy E_γ.

N is the net peak area under the specific peak corrected for the background at energy E_γ.

A is the activity in (Bq) of standard source at the measured time.

$I_\gamma(E_\gamma)$ is the abundance at energy E_γ.

t is the time of measurement (3600sec).

The efficiency of the detector is related to energy by the expression:

$$\begin{aligned} ln\varepsilon &= 5.576e1-1.14e2*ln(E) \\ &+5.855e1*ln(E)\wedge 2-1.286e1*ln(E)\wedge 3 \\ &+1.296*ln(E)\wedge 4-4.949e-2*ln(E)\wedge 5 \end{aligned} \quad (2)$$

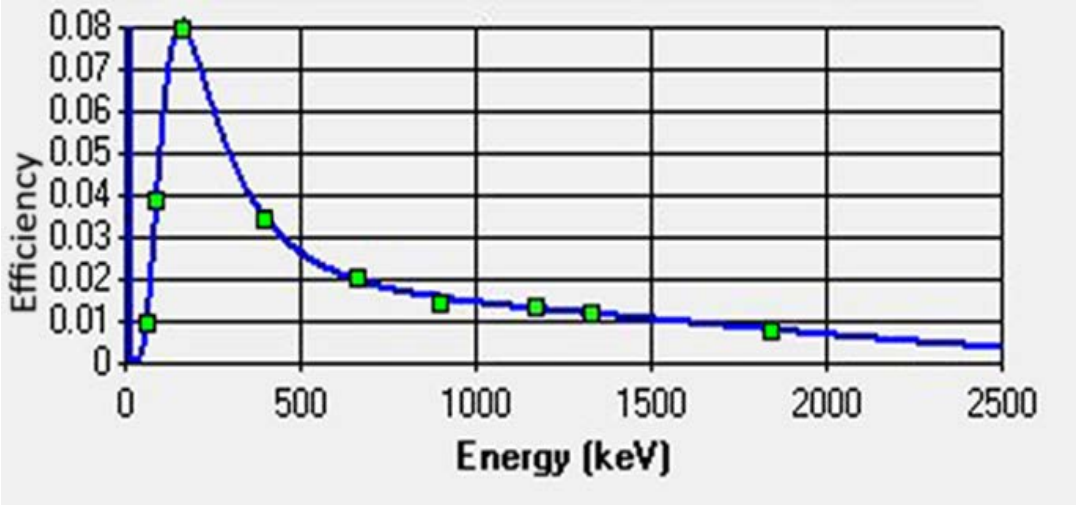

Figure 2. The efficiency calibration curve for the (HPGe) detector

2.5. Specific Activity Calculation:

Some radionuclides of the natural decay chains (e.g. ^{238}U, ^{232}Th, and ^{226}Ra) cannot be determined directly by gamma spectrometry but only by measuring their daughter radionuclides. In these cases, it is necessary to ensure that there is equilibrium between the parent radionuclide and the daughter radionuclides being measured.

Several transitions from decays of shorter-lived radionuclides in the ^{238}U decay chain, such as ^{226}Ra, ^{214}Pb and ^{214}Bi (which can be thought of as ^{226}Ra indicators), were used to estimate the weighted mean of activity concentration of ^{226}Ra. The activity concentration of ^{232}Th is determined using gamma-ray transitions associated with the decay of ^{228}Ac. The gamma-ray peaks associated with decays from ^{40}K at 1461 keV, are used to determine the activity concentrations for these nuclei. The lines suitable for evaluation have been compiled In Table 1.

Table 1. Selected gamma lines for the determination of natural radionuclides [8]

Radionuclide determined	Radionuclide measured	Energy E_γ keV	Emission probability I_E %
^{226}Ra	^{226}Ra	186.10	3.51
	^{214}Bi	609.31 1120.29 1238.11 1764.49	44.6 14.70 5.94 15.10
^{232}Th	^{228}Ac	209.25 338.32 911.60 968.97	3.89 11.27 25.80 15.8
^{40}K	^{40}K	1460.83	10.67

The specific activity, in terms of the activity concentration, is defined as the activity per unit mass of the sample. The specific activity of individual radionuclides in soil samples is given by the following equation [4]:

$$A(Bq/kg) = \frac{N}{\varepsilon(E_\gamma) I(E_\gamma) t_c m} \quad (3)$$

Where
N = the corrected net peak area of the corresponding full-energy peak

$$N = N_S - N_B \quad (4)$$

N_S = the net peak area in the sample spectrum,
N_B = the corresponding net peak area in the background spectrum,
$\varepsilon(E_\gamma)$ = the counting efficiency of the specific nuclide's energy,
$I(E_\gamma)$ = the absolute transition probability by gamma decay through the selected energy (E_γ),
t_c = the counting live-time of the sample spectrum collection in seconds,
m = the mass (kg) of the measured sample.

2.6. Absorbed Dose Rate

The absorbed dose rate (D_γ) due to gamma radiation of naturally occurring radionuclides ^{238}U, ^{232}Th and ^{40}K, were calculated on guidelines provided by UNSCEAR [2].

$$D_\gamma\,(\text{nGy/h}) = 0.462A_{Ra} + 0.621A_{Th} + 0.042A_K \quad (5)$$

Where the factors (0.462, 0.621 and 0.042) are the conversion factors for naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K, respectively. The average of (D_γ) in air (1m) from terrestrial sources of gamma radiation in soil is estimated as (55 nGy/h) [9].

2.7. Radium Equivalent Activity (Ra_{eq})

Due to the non-uniformity in the distribution of natural radionuclides in the soil samples, the actual activity level of ^{238}U, ^{232}Th and ^{40}K in the samples can be evaluated by means of a common radiological index named the radium equivalent activity (Ra_{eq}). Radium equivalent activity (Ra_{eq}) is used to assess the hazards associated with materials that contain ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg [2], which is determined by assuming that 370 Bq/kg of ^{226}Ra or 260 Bq/kg of ^{232}Th or 4810 Bq/kg of ^{40}K produce the same γ dose rate [7,10]. The Ra_{eq} of a sample in (Bq/kg) can be achieved using the following relation [7]:

$$Ra_{eq}\,(\text{Bq/kg}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (6)$$

Where, A_{Ra}, A_{Th} and A_K are the specific activities concentrations (Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The published maximal admissible (permissible) Ra_{eq} is 370 Bq/kg [2]. Which corresponds to an effective dose of 1 mSv for the general public [11].

2.8. External Hazard Index

The external hazard index is an evaluation of the hazard of the natural gamma radiation [12]. The prime objective of this index is to limit the radiation dose to the permissible dose equivalent limit of $1mSvy^{-1}$ [13,14]. In order to evaluate this index, a model proposed by Beretka and Mathew in 1985 [10]. This assumes the following relation:

$$H_{ex} = \left(\frac{A_{Ra226}}{370}\right) + \left(\frac{A_{Th232}}{259}\right) + \left(\frac{A_{K40}}{4810}\right) \le 1. \quad (7)$$

This model takes into account that the external hazard which is caused by gamma-rays corresponds to a maximum radium-equivalent activity of 370 Bq/kg for the material [7,15]. In order to keep the radiation hazard insignificant, the value of external hazard index must not exceed the limit of unity [6].

3. Results and Discussion

The results can be summarized as:
(i) Activity concentrations, and (ii) Radiological indices.

3.1. Activity Concentrations

Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides in the Fuel Fabrication Facility soil samples were determined by equation (3) and the results have been shown in Table 2. The activity concentration of ^{226}Ra in the soil ranged from 4.7±0.5 (IFFF-8) to 25.1±1.6 (IFFF-2) $Bq.kg^{-1}$ with mean 15.78±1.16 $Bq.kg^{-1}$, ^{232}Th ranged from 3.1±0.6 (IFFF-8) to 22.9±1.2 (IFFF-5) $Bq.kg^{-1}$ with mean 14.09±1.33 $Bq.kg^{-1}$, and ^{40}K ranged from 75.7±7.2 (IFFF-8) to 475.5±25.8 (IFFF-2) $Bq.kg^{-1}$ with mean 306.42±18.1 $Bq.kg^{-1}$, respectively. The average activity concentrations of terrestrial radionuclides ^{226}Ra, ^{232}Th and ^{40}K are within the world wide average concentrations of these radionuclides reported by UNSCEAR (2000) as 35, 30 and 370 Bq kg^{-1}, respectively.

Table 2. The activity concentrations values for different soil samples

Sample code	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
IFFF-1	18.3±1.2	19.7±1.2	380.9±21.9
IFFF-2	25.1±1.6	18.9±2	475.5±25.8
IFFF-3	21.8±1.6	20.2±2.4	383.7±22.7
IFFF-4	12.5±0.8	13±0.9	270.1±15.7
IFFF-5	19.6±0.9	22.9±1.2	306.6±16.5
IFFF-6	11.7±1.1	19.8±2.3	303.6±19.9
IFFF-7	12.4±0.7	10±0.8	262.4±14.3
IFFF-8	4.7±0.5	3.1±0.6	75.7±7.2
IFFF-9	16.3±1.5	13.3±1.9	304.9±17.8
IFFF-10	15.4±1.7	<MDA	300.8±19.2
Min.	4.7±0.5	3.1±0.6	75.7±7.2
Max.	25.1±1.6	22.9±1.2	475.5±25.8
Average	15.78±1.16	14.09±1.33	306.42±18.1
World mean value	35	30	370

3.2. Radiological Indices

In order to assess the health effects, the radiation hazards such as absorbed dose rate (Dγ), radium equivalent activity (Ra_{eq}), and external hazard index (H_{ex}) have been calculated from the activity of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides using the equations (5), (6) and (7), respectively and the values have shown in Table 3.

The Table 2 shows that the absorbed dose rate in air due to terrestrial gamma rays at 1m above the ground were calculated for ^{226}Ra, ^{332}Th and ^{40}K and ranged from 7.28±0.91 to 43.3±3.06 nGyh^{-1} with mean of 28.90±2.12 nGyh^{-1} which is lower than the world average value of 55 nGyh^{-1} [9].

Table 3. Absorbed dose rate, Radium equivalent activity and External hazard index

Sample code	Absorbed dose rate (D_γ) (nGy/h)	Radium equivalent activity (Ra_{eq}) (Bq.kg^{-1})	External hazard index (H_{ex})
IFFF-1	36.59±2.4	75.6±5	0.2±0.01
IFFF-2	43.3±3.06	88.74±6.45	0.24±0.02
IFFF-3	38.73±3.18	80.23±6.78	0.22±0.02
IFFF-4	25.19±1.59	51.89±3.3	0.14±0.01
IFFF-5	36.16±1.85	75.97±3.89	0.21± 0.01
IFFF-6	30.45±2.77	63.39±5.92	0.17±0.02
IFFF-7	22.96±1.42	46.9±2.95	0.13±0.01
IFFF-8	7.28±0.91	14.96±1.91	0.04±0.01
IFFF-9	28.6±2.62	58.8±5.59	0.16±0.02
IFFF-10	19.75±1.59	38.56±3.18	0.1±0.01
Min.	7.28±0.91	14.96±1.91	0.04±0.01
Max.	43.3±3.06	88.74±6.45	0.24±0.02
Average	28.90±2.12	59.52±4.45	0.16±0.012
Global limit	55	370	≤ 1

4. Conclusions

This study shows that the average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil samples were found to be 15.78±1.16, 14.09±1.33 and 306.42±18.1 Bqkg^{-1}, respectively. The results obtained for the corresponding radionuclides are below the worldwide average values of 35, 30, and 400 Bqkg^{-1}, respectively. The average absorbed dose rate in air (Dγ), the average radium equivalent activity (Ra_{eq}) and the average external hazard index (H_{ex}) were determined as 28.90±2.12 nGyh^{-1}, 59.52±4.45 Bqkg^{-1} and 0.16±0.012, respectively, which are below the permissible limit. However, this data may provide a general background level for the area studied and may also serve as a guideline and a baseline data for future measurement and assessment of possible radiological risks to human health in Al-Tuwaitha site.

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