Radiological risk assessment of fuel fabrication facility at Al- Tuwaitha nuclear site

Yousif M. Zayir Al-Bakhat[1], Altaf Abdul Majeed Salman[2], Zainab M. Jbara[1]

[1]Radiation & Nuclear Safety Directorate, Ministry of Science & Technology

[2]Department of Physics, College of Science for Women, Baghdad University

E-mail: yousif_zayir@yahoo.com

Abstract

The specific activity of 29 soil samples collected from Fuel Fabrication Facility FFF at AL-Tuwaitha site, 20 km south of Baghdad were determined using HPGe detector in a low background configuration, it's relative efficiency of 40%, and resolution of 2keV for the 1332 keV gamma ray emission of ^{60}Co. The range of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were between (12.56-31.96), (10.2-18.4) and (47.47-402.1) Bq/kg respectively. In order to assess any radiological hazard to human health, the absorbed gamma dose rate D in air at 1m above the ground surface was calculated in the range (18.87 to 36.46) nGy/h; the outdoor annual effective dose equivalent *AEDE* was evaluated to vary from 0.0039 to 0.0076 mSv/y with the mean value 0.0059 mSv/y, this value comparable to the worldwide effective dose 0.48mSv/y. The Radium Equivalent Ra_{eq} for all samples were evaluated and were lower than the accepted safety limit value of 370 Bq/kg. The results indicated that the radiation of hazards from primordial radionudlides in all samples in this study is not significant.

Key words

Ra_{eq} activities, Annual effective dose, External hazard index.

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تقييم المخاطر الإشعاعية لمنشأة تصنيع الوقود في موقع التويثة النووي

يوسف محسن زاير[1]، ألطاف عبد المجيد سلمان[2] ، زينب موسى جبارة[1]

[1]مديرية السلامة الإشعاعية والنووية، وزارة العلوم والتكنولوجيا

[2]قسم الفيزياء، كلية العلوم للبنات، جامعة بغداد

الخلاصة

النشاط الإشعاعي الطبيعي لـ 29 نموذج تربة جمعت من موقع تصنيع الوقود في موقع التويثة النووي FFF الواقع على بعد 20 km جنوب بغداد، تم الحصول عليه من استخدام مطياف أشعة كاما ذو كاشف الجرمانيوم عالي النقاوة HPGe بترتيب منخفض للخلفية الإشعاعية و بكفاءة 40% وقدرة تحليل 2keV لنظير الكوبلت ^{60}Co عند الطاقة 1332 keV. كانت تراكيز النويدات المشعة (K^{40},Th^{232},Ra^{226}) تتراوح بين (12.56-31.96)، (10.2-18.4)، (47.47-402.1) Bq/kg على التوالي. أما بخصوص تقييم المخاطر الإشعاعية على الصحة فان معدل جرعة كاما الممتصة D في الهواء وعلى ارتفاع 1m فوق مستوى سطح الأرض قد تم حسابه ليكون ضمن المدى (18.87-36.46 nGy/h) اما الجرعة السنوية المؤثرة (AEDE) فقد كانت ضمن المدى 0.0039-0.0076 mSv/y، وبمعدل، 0.0059 mSv/y، هذه القيم تم مقارنتها مع الجرعة المؤثرة عالميا والتي تساوي (0.48mSv/y). أما بالنسبة لمكافئ الراديوم Ra_{eq} لجميع النماذج كان اقل من حد الأمان المقبول والذي قيمته (370 Bq/kg)، هذا يعني أن النتائج التي حصلنا عليها تشير إن المخاطر الإشعاعية الناتجة من النويدات المشعة الأساسية والتي جمعت من هذا الموقع هي مخاطر غير مؤثرة.

Introduction

The radioactive waste resulting from the preceding nuclear activities in Iraq has the potential to cause significant radiological issues to the environment and to the public living in the neighborhood of these nuclear sites. The destruction of the former nuclear facilities during the 1991 Gulf war, and the looting of the sites and facilities during the 2003 events, aggravated the problem. As a result of these events, many of these nuclear facilities have lost their containment of the radioactive material and it now has an increased potential to be dispersed into the environment. All sites and facilities require decommissioning in order to ensure both radiological and non-radiological safety [1]. Despite the long history of nuclear programs at Al-Tuwaitha no significant radioactive contamination as a result of normal operations has been officially reported for the site or surrounding communities. Radionuclide's are present in the environment and within the remaining structures [2]. Location of these facilities was shown in Fig.1.

Fig.1: Map of Al-Tuwaitha nuclear sites facilities.

Radioactive elements are generally classified into two general types: naturally occurring and artificially produced radioactive isotopes. Natural and artificial radioactivity is explained in detail in the next section. Terrestrial radiation arising from the Earth's crust and building materials gives rise to the external exposure and inhalation or ingestion of natural radionuclides. The most important source of external radiation exposure is due to gamma-rays emitted from the ^{238}U and ^{232}Th decay chains and ^{40}K isotope. The radiological implication of these radionuclides is due to the gamma-ray exposure of the body and irradiation of the lung tissue from inhalation of radon and its daughters [3].

Artificial radioactivity arises mainly from discarded radioactive sources, radioactive wastes and radioactive fall-out [4]. Human activities like mining and milling of mineral ores, ore processing and enrichment, nuclear fuel fabrication and handling of the fuel cycle tail end products have contributed to the increased concentration of some of radionuclides in the environment [5]. The intensity of the terrestrial natural radioactivity varies by an order of magnitude for different regions of the world due to geological and environmental factors. The variations in the abundance and distribution of the primordial radionuclides in the environment account for the spatial variations in the natural gamma radioactivity of such environment. In the terrestrial environment, soil is the main reservoir of radionuclides and it acts as a medium of migration for the transfer of these radionuclides to the biosphere. Radionuclides are initially deposited on the upper surface of the soil, but they are quickly weather into the first few centimeters of the soil [6]. Among the radionuclides produced artificially and released into the marine environment, ^{137}Cs isotope is extremely dangerous [7].

Materials and methods

1. The studied area

Fig.1 shows the geographic location of the Fuel Fabrication Facility (FFF) at Al-Tuwaitha site was a part of the nuclear Italian complex, GPS Coordinates (North= 33°11.462″, East=44°30.723″) which consist:
1-The Italian radioisotopes production laboratory and 2 -The Fuel Fabrication Facility FFF.

The Fuel Fabrication Facility covered an area of about 32000 m^2.It was operated in 1981 and destroyed in 1991 through the second Gulf war. It was used to purify yellow-cake to produce U_3O_8, UO_2, UO_3 and metallic uranium. Some fuel elements using natural fuel were also produced at this facility, which were later irradiated at IRT 5000 for the purpose of radiochemistry research [8].

2. Samples collection and preparation

Twenty nine soil samples of 1 kg within the consideration area were collected as uniformly as possible, from a depth of 20 cm they were collected at average intervals nearly 50 m between locations .The soil samples, initially filled into polyethylene bags separately for respective points in equal measures and labeled accordingly for benchmarking purposes. Following thorough drying the soil were pulverized to fine power and packed to fill cylindrical containers of height 7 cm by 6 cm in diameter which is the same as the geometry of the counting detector. Each container accommodated approximately 500 g of sample. Samples were sealed and stored for four weeks to achieve secular equilibrium between radium and its

progeny. Each sample was measured for a time period of 3600 seconds.

3. Measurement of natural radioactivity

Gamma transitions of 1461 keV for ^{40}K , 186 keV and 609 keV of ^{214}Bi for ^{226}Ra, 338, 463, 911, 968 keV for ^{228}Ac, 727 keV for ^{212}Bi, 238 keV for ^{212}Pb were used for the laboratory measurement of activity concentration potassium, radium and thorium. The samples were counted for a period of 72000 seconds. The concentrations of radionuclides are calculated using the following equation [9]:

$$A(E_\gamma) = \frac{N}{t \times I_\gamma(E_\gamma) \times \varepsilon(E_\gamma) \times m} \quad (1)$$

where

N = net peak area under the specific peak corrected for the background at energy(E_γ).

t= live time of the sample spectrum collection in seconds

$I_\gamma(E_\gamma)$ = abundance at energy (E_γ)

$\varepsilon(E_\gamma)$= efficiency at photo peak energy

m = mass (kg) of the measured sample

4. Absorbed dose rate in air (*D*)

In order to assess any radiological hazard, the exposure to radiation arising from radionuclides present in soil can be determined in terms of many parameters. A direct connection between radioactivity concentrations of natural radionuclides and their exposure is known as the absorbed dose rate in the air at 1 meter above the ground surface. The mean activity concentrations of ^{226}Ra (^{238}U), ^{232}Th, and ^{40}K (Bq/kg) in the soil samples are used to calculate the absorbed dose rate given by the following formula: [9, 10]

$$D \text{ (nGy/h)} = (0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K) \quad (2)$$

where D is the absorbed dose rate in nGy/h, A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K, respectively. The dose coefficients in units of nGy/h per Bq/kg were taken from the UNSCEAR 2000 report [11].

5. Annual Effective Dose Equivalent *AEDE*

The absorbed dose rate in air at 1 meter above the ground surface does not directly provide the radiological risk to which an individual is exposed [12]. The absorbed dose can be considered in terms of the annual effective dose equivalent from outdoor terrestrial gamma radiation which is converted from the absorbed dose by taking into account two factors, namely the conversion coefficient from absorbed dose in air to effective dose and the outdoor occupancy factor. The annual effective dose equivalent can be estimated using the following formula [11, 13]:

$$AEDE_{\text{out}} \text{ (mSv/y)} = \text{D (nGy/h) x 8760 h x 0.2 x 0.7 x } 10^{-6} \text{ Sv/Gy} \quad (3)$$

The values of those parameters used in the UNSCEAR report (2000) are 0.7 Sv/Gy for the conversion coefficient from absorbed dose in air to effective dose received by adults and (0.2) for the outdoor occupancy factor [6].In the present work we take the time 1440 h instead of 8760 h according to the our condition in the selected site.

6. Radium equivalent activity (Ra_{eq})

Due to a non uniform distribution of natural radionuclides in the soil samples, the actual activity level of ^{226}Ra, ^{232}Th and ^{40}Kin the samples can be evaluated by means of a common radiological index named the radium equivalent activity (Ra_{eq}) . It is the most widely used index to assess the radiation hazards and can be calculated

using Eq.(4) given by Beretka and Mathew [13]. This estimates that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same gamma-ray dose rate [14, 15, 16].

$$Ra_{eq}(\text{Bq/kg}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (4)$$

The permissible maximum value of the radium equivalent activity is 370 Bq/kg [11], which corresponds to an effective dose of 1 mSv for the general public [17].

7. External and internal (*Hex*, *Hin*) hazard index

To limit the radiation exposure attributable to natural radionuclides in the samples to the permissible dose equivalent limit of 1 mSv/y, the external hazard index based on a criterion have been introduced using a model proposed by Krieger (1981) which is given by the equations [15, 16, 18].

$$H_{ex} = \left[\frac{A_{Ra}}{370}\right] + \left[\frac{A_{Th}}{259}\right] + \left[\frac{A_K}{4810}\right] \leq 1 \quad (5)$$

From above equation when the value of external index is not exceed the limit of unity that is mean the radiation hazard insignificant, the maximum value of external index equal to unity correspond to the upper limit of radium equivalent activity 370 Bq/kg [16].

$$H_{in} = \left[\frac{A_{Ra}}{185}\right] + \left[\frac{A_{Th}}{259}\right] + \left[\frac{A_K}{4810}\right] \leq 1 \quad (6)$$

The value of H_{in} must also be less than unity.

Results and discussion

1. Spesific activity measurements

The specific activity of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg for soil samples were carried out using a gamma-ray spectrometry system with a high purity germanium (HPGe) detector in a low background configuration, and a relative efficiency of 40%, and resolution of 2keV for the 1332 keV gamma ray emission of ^{60}Co.

Table 1 illustrates the specific activities of natural radionuclides ^{226}Ra,^{232}Th, and ^{40}K in the samples collected from (FFF) nuclear sites. Activity of ^{226}Ra, collectively for all 29 samples, found to be ranged from 12.56±0.32 to 31.96±0.016 Bq/kg with an average value 17.120±4.114 Bq/kg. The ^{232}Th activity ranged from 10.2±0.58 to 18.4±0.09 Bq/kg with an average value of 14.581±2.019 Bq/kg. While the activity of ^{40}K ranged from 47.47±6.06 to 402.1±15.01 Bq/kg with an average of 285.40±61.75 Bq/kg.

Table 2 shows the range and average values (with their standard deviation) of radium equivalent activity Ra_{eq}, absorbed dose rate in air outdoors D, annual effective dose equivalent AEDE, external hazard index H_{ex} and internal hazard index H_{in} of ^{226}Ra, ^{232}Th series and ^{40}Kin surface soils of FFF site.

Table 1: Illustrated the measurements of activity concentrations in Bq/kg.

No.	Sample Code	Activity concentration Bq/kg ^{226}Ra	Activity concentration Bq/kg ^{232}Th	Activity concentration Bq/kg ^{40}K
1	S1	17.7 ±0.51	14.79±0.61	367.9±0.31
2	S2	15.34±0.66	11.21±1.22	279.6±0.65
3	S3	18.24±0.21	14.1±0.53	277.3±0.55
4	S4	12.56±0.32	15.13±1.01	281.9±12.1
5	S5	16.33±0.85	12.89±0.21	207.3±10.2
6	S6	16.81±0.52	13.2±2.01	332.1±11.68
7	S7	20.35±2.01	15.77±3.05	257.7±9.12
8	S8	14.06±5.02	17.21±0.04	47.47±6.06
9	S9	20.62±0.33	13.09±0.51	402.1±15.01
10	S10	18.5±0.06	10.2±0.58	242.7±8.21
11	S11	14.46±0.05	11.72±0.66	325.1±14.12
12	S13	20.26±0.06	13.42±0.09	240.2±14.23
13	S15	22.2±0.078	14.95±1.09	337.6±1.45
14	S16	17.3±0.056	15.4±2.03	240.9±6.54
15	S17	20.15±0.022	13.9±2.03	299.2±9.88
16	S22	19.1±0.09	15.7±0.31	311.8±0.02
17	S23	14.4±0.21	17.7±0.65	333.5±1.03
18	S24	14.84±0.33	15.54±0.06	309±0.09
19	S25	15.6±0.34	18.4±0.09	330.7±0.06
20	S27	12.90±0.11	13.4±0.54	280.02±0.07
21	S28	14.92±0.52	15.47±0.36	323.2±0.12
22	S30	13.96±0.33	17.24±0.84	297.3±0.09
23	S31	13.660.41	14.28±0.24	266.8±0.84
24	S32	13.3±0.08	11.26±0.051	248.2±0.32
25	S33	31.96±0.016	16.84±0.22	276.35±0.6
26	S35	23.55±0.22	16.77±0.41	304.6±0.008
27	S36	14.55±0.38	14.15±0.51	276.1±0.002
28	S37	13.63±0.52	14.41±0.66	270.9±0.008
29	S46	15.26±0.44	14.72±0.31	309.1±0.62
Average ±S.D.		**17.120±4.114**	**14.581±2.019**	**285.40±61.75**
World wide average concentrations [11]		35	30	400

S.D.=standard deviation.

Table 2: The experimental results of radium equivalent activity Ra_{eq}, absorbed dose rate D, annual effective dose equivalent AEDE, external H_{ex} and Internal H_{in} radiation hazard indices.

Sample code	Ra_{eq}(Bq/kg)	D(nGy/h)	AEDE(mSv/y)	H_{ex}	H_{in}
S1	67.18	32.45	0.00681	0.1814	0.2293
S2	52.9	25.52	0.00536	0.1429	0.1843
S3	59.76	28.51	0.00599	0.1614	0.2107
S4	55.9	26.7	0.00561	0.151	0.1849
S5	50.72	23.97	0.00503	0.137	0.1811
S6	61.25	29.58	0.0062	0.1654	0.2108
S7	62.74	29.67	0.00623	0.1695	0.2245
S8	42.33	18.87	0.00396	0.1143	0.1523
S9	70.3	34.2	0.00718	0.1899	0.2456
S10	51.77	24.83	0.00521	0.1398	0.1898
S11	56.25	27.32	0.00574	0.1519	0.191
S13	57.95	27.48	0.00577	0.1565	0.2113
S15	69.57	33.36	0.00701	0.1879	0.2479
S16	57.87	27.34	0.0057	0.1563	0.2031
S17	63.07	30.18	0.0063	0.1703	0.2248
S22	65.56	31.31	0.00657	0.1771	0.2287
S23	65.39	31.25	0.00656	0.1766	0.2155
S24	60.86	29.13	0.00612	0.1643	0.2045
S25	67.38	32.11	0.0067	0.182	0.2241
S27	53.62	25.73	0.0054	0.1448	0.1797
S28	61.93	29.71	0.0062	0.1672	0.2076
S30	61.51	29.26	0.0061	0.1661	0.2038
S31	54.62	26.06	0.0054	0.1475	0.1844
S32	48.51	23.3	0.0048	0.131	0.167
S33	77.32	36.46	0.00766	0.2089	0.2952
S35	70.99	33.71	0.0070	0.1917	0.2554
S36	56.04	26.78	0.0056	0.1514	0.1907
S37	55.1	26.3	0.0055	0.1488	0.1856
S46	60.11	28.83	0.0060	0.1623	0.2036
Average ±S.D.	59.94±7.57	28.61±3.72	0.0059±0.00078	0.161±0.02	0.208±0.029
World Median Value*	<370	57	0.48 mSv/y	≤1	≤1

*= UNSCEAR−2000.

The estimated radium equivalent activity of surface soils were varied from 42.33 to 77.32 Bq/kg with an average value and standard deviation of 59.94±7.57 Bq/ kg were less than the permissible level of 370 Bq/ kg recommended by UNSCEAR-2000 [11].

The absorbed dose rates were calculated for each sample at 1 meter above ground surface in air outdoors using Eq. (2). Gamma absorbed dose rates ranged between 18.87 and 36.46 nGy/h with an average value with standard deviation of 28.61±3.72 nGy/h. This indicates that the absorbed dose rate from terrestrial

radiation in air outdoors in the investigated area were less than the permissible limit recommended by UNSCEAR-2000. The annual effective dose equivalent due to the specific activity levels of ^{226}Ra, ^{232}Th and ^{40}K were calculated according to Eq. (3) and found to be vary from 0.00396 to 0.00766 mSv/y with average value and standard deviation of 0.0059±0.00078 mSv/y. The range and average values for the annual effective dose rate were less than the worldwide median value recommended by, UNSCEAR− 2000. The external and internal radiation hazard indices were calculated by Eq. (5) and Eq.(6) and found to be with an average value and standard deviation of 0.161±0.02 and with an average value and standard deviation of 0.208±0.029 respectively from these results we can see that the radiation hazard indices not to exceed the permissible limits of 1.

Conclusions

The conclusion studying the natural radionuclide specific activity in surface soils of randomly selected in Fuel Fabrication Facility FFF site has been reported. The mean activity of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K were found to be 17.120, 14.581 and 285.40 Bq/kg, respectively. Despite the fluctuation in the measurements of the specific activity of each natural radionuclide ^{226}Ra (^{238}U), ^{232}Th and ^{40}K in the studied soil samples, the data are found to be normal and lower than the world average of (35, 30 and 420) Bq/kg respectively. The results for dose rate in air outdoors from terrestrial gamma rays are less than the average world values. The annual effective dose was found to be 0.0059 mSv/y, which is well below the permissible dose equivalent. Also the external and internal radiation hazards indices less than unity that is mean there is no radiation risk in this site.

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