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Assessment of Radioactivity Levels for the Fuel Fabrication Facility at Al-tuwaitha Nuclear Site (Iraq)

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Abstract: The activity concentrations of twenty soil samples collected from contaminated locations of Fuel Fabrication Facility (FFF) at Al- Tuwaitha nuclear site, Iraq, were determined by using spectroscopy with a high purity germanium detector. The range of activity concentrations of ^{238}U (^{234m}Pa), ^{235}U, ^{232}Th (^{228}Ac), and ^{40}K in the soil from the studied areas varies from (177.74±60) to (375777.5±6895) $Bqkg^{-1}$, (7.4±0.5) to (20954±407.5) $Bqkg^{-1}$, (7.06±0.22) to (20.45±0.33) $Bqkg^{1}$ and from (47.5±5.3) to (402.2±15.5) Bq kg^{-1} respectively. The isotopic mass and activity ratio of $^{235}U/^{238}U$ were calculated to identify kind of uranium, depleted or natural. To assess the radio logical risk for this site, RESRAD program was used to calculate total dose from all pathways (external, inhalation and soil ingestion) of exposure, the value of total dose was 2mSv/yr. Laboratory results indicated that the FFF was contaminated with ^{234m}Pa and ^{235}U nuclides in excess of the IAEA limits for exemption from regulatory control, and indicating that the decommissioning operations for the FFF must be subjected to regulatory control and safety surveillance to ensure adequate protection of the operators, public and the environment.

Keywords: Nuclides Activity Concentration, Mass and Activity Isotopic Ratio $^{235}u/^{238}u$, Risk Assessment, Fuel Fabrication Facility

1. Introduction

The use of radioactive materials has made significant impact in various areas and lead to changes in human practices [1]. In fact, no one can escape from being exposed to ionizing radiation. However, the amount of exposure differs depending on human practices and surroundings [2].

In recent years, it recognized widely became that there is a large number of sites in different countries of the world that have become contaminated by radiation as a result of the activities of the nuclear fuel cycle; nuclear weapons programs; the use of radioisotopes in medicine, research and industry; accidents; and so on. For some sites, localized contamination has occurred to some extent as a result of the operations or processes, for example, contamination from factory operations radium luminescence or burial or disposal of specific industrial radioactive materials. For other cases, contamination on a wide range with radioactive material occurred following dismantling activities [3]. The existence of such a radioactivity of these activities, improved the natural man or man-made, can pose a risk to human health or the environment. Therefore, characterization and proper treatment may be a mandatory requirement in order to reduce the radiological risk to acceptable levels [4]. A number of sites in Iraq have some degree of radiological contamination and require decommissioning and remediation in order to ensure radiological safety. Many of these sites in Iraq are located at the nuclear research center at A1-Tuwaitha. The International Atomic Energy Agency (IAEA) Board of Governors has approved a project to assist the Government of Iraq in the evaluation and decommissioning of former facilities that used radioactive materials. There are a number of sites in Iraq which have been used for nuclear activities and which contain potentially significant amounts of radioactive material. Many of these sites suffered substantial heavy damage during the Gulf Wars and several have been subject to looting of materials and equipment as a consequence of the challenging security

situation in the country. One of these sites is Italian Fuel Fabrication Facility (FFF) located at the site of the nuclear Tuwaitha 20 km south of Baghdad [5].

2. Method and Measure

2.1. Sample Collection

In order to estimate the levels of radioactivity concentration in Fuel Fabrication Facility (FFF) at Al-Tuwaitha nuclear site, 20 surface soil samples have been collected. The samples have taken at 25 cm depth from the top surface soil layer to make approximately 1 kg weight per sample, using hand auger and coring tool, each soil sample is filled into secure polyethylene bag to prevent cross contamination and sent to the laboratory.

2.2. Sample Preparation

To prepare the soil samples for measuring activity concentrations in the laboratory, at first, these samples have placed in an oven at a temperature of 80 ^{0}C for 2 hours, especially samples that were collected after the rain, thus make ensure complete removal of any residual humidity. The dried samples have crushed into a downy powder and passed through a standard 75µm mesh size. The prepared samples have filled into 550 ml Marinelli beakers which then closed well and labeled with the plastic tape to prevent outflow of airborne ^{220}Rn and ^{222}Rn from the samples. Collected samples were weighed and stored for one month in order to maintain and achieve radioactive secular equilibrium between ^{226}Ra and ^{228}Ac and their short-lived progeny (>7 half-lives of ^{222}Rn and ^{220}Rn). The information of each sample is documented separately in prepared labels, attached with each sample bag.

2.3. Instrumentations

Gamma spectrometer (Canberra) system has used to measureand analyze samples using vertical high purity germanium (HpGe) detector of efficiency 40%, and resolution (2.0 keV). Based on the measurement of 1332 keV gamma ray photo peak of ^{60}Cosource and Multichannel analyzer (MCA) with 8192 channel was used [13]. A library of radio nuclides contained the energy of the characteristic gamma emissions of each nuclide has analyzed and their corresponding emission probabilities were built from the data supplied in the software (Genie-2000) [4]. The specific activity of individual radio nuclides in soil samples is given by the following equation [4, 5].

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

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

t: the live time of the sample spectrum collection in seconds.

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

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

m: the mass (kg) of the measured sample

2.4. Determination of Isotopic Activity Ratio $^{235}U/^{238}U$

The ^{238}U activity concentration can be determined using the 1001.03 keV (0.837%) gamma line associated with^{234m}Pa[6]. By evaluating the activity concentration of ^{238}U (^{234m}Pa) and ^{235}U activity, the ^{235}U / ^{238}U activity ratio can be determined for each sample. Any differences from the expected natural isotopic ratio for uranium may be due to relative distribution of the specific radioisotopes(decay daughter) in a particular geological environment[7]. Natural Uranium (U) principally consists of three isotopes, primordial^{238} U ($t_{1/2}$ = 4.47×10^9 y) and ^{235}U ($t_{1/2}$ = 7.04×10^8 y), which are parent members of a natural radioactivedecay series, and ^{234}U ($t_{1/2}$ = 2.45 × 10^5 y), which is a member of the ^{238}U decay chain. For natural U, the ^{235}U/^{238}U activity ratio has a constant value 0.046 and mass ratio has 0.0072 (Table 1), while the ^{234}U/^{238}U activity ratio is variable as a onsequence of decay chain disequilibrium that arises from preferential transfer of ^{234}U to surface and groundwater [8, 9]. This disequilibrium results in pronounced ^{234}U /^{238}U activity ratio variations, but for soils the commonly observed range is 0.8–1.2. Depleted Uranium (DU) is a byproduct of U enrichment processes, whereby the fissile isotope ^{235}U is preferentially concentrated for the production of nuclear fuel research reactors. The enrichment processes, e.g. gas centrifugation or gaseous diffusion, also separate ^{234}U from ^{238}U, leaving a waste material (DU) which is depleted with respect to both ^{234}U and ^{235}U [10].

Table 1. *Typical Isotope abundance and activity ratios in natural and depleted uranium [12].*

	Atomic abundance (%)	**Activity ratio**				**Mass ratio**	
	238U	235U	234U	235U: 238U	234U: 238U	235U: 238U	234U:238 U
Natural U	99.27	0.72	5.5×10^{-3}	7.2×10^{-3}	5.5×10^{-5}	0.046	0.8- 1.2
DU	99.80	0.20	9×10^{-4}	2.0×10^{-3}	9×10^{-6}	0.013	0.193

As 234U does not emit suitable photons for gamma spectrometry, the detection of DU contamination by gamma spectrometry is mainly based on the determination of the 235U/238U ratio. Furthermore, since 238U does not emit significant photons that can be used for its gamma spectroscopic determination, it is usually determined through its daughter products in equilibrium, namely ^{234}Th and ^{234m}Pa [11]. For calculating the mass ratio ^{235}U / ^{238}U, e;

$$\mathbf{e} = \frac{\mathrm{m}(235)}{\mathrm{m}(235)+\mathrm{m}(238)} \times 100 \quad (2)$$

where *m*(235) and *m*(238) are the masses of ^{235}U and ^{238}U respectively. Generally, the mass of ^{234}U is very small compared to the masses of the two other uranium isotopes

and it was neglected in this formula. Considering the fact that both radio nuclides are radioactive, the enrichment can be expressed as a function of the activity values of ^{238}U and ^{235}U. Starting from the basic formula:

$$A = \frac{\ln 2}{T_{1/2}} \times N \quad (3)$$

Where A is the activity, N- is the number of radioactive nuclei and T1/2 the half-life, the masses of the two isotopes are:

$$m(235) = \frac{A(235) \times 235. \times T_{1/2}(235)}{N_A. \times \ln 2} \quad (4)$$

$$m(238) = \frac{A(238) \times .238. \times T_{1/2(238)}}{N_A. \times \ln 2} \quad (5)$$

where A (235) and A(238) are the activity values of ^{235}U and ^{238}U respectively;$T_{1/2}(235)$ =7.04×10^8years and $T_{1/2}(238)$ = 4.47×10^9years are their half-livesand N_A is the Avogadro's constant. Using the formulae (2), (4) and (5), therefore the mass ratio equation can be shown as:

$$e = \frac{1}{1+\frac{A(238)}{A(235)} \times \frac{T_{1/2(238)}}{T_{1/2(235)}} \times \frac{238}{235}} . 100 \quad (6)$$

3. Results and Discussion

Table 1 shows the results obtained for the levels of specific activity concentrations for radionuclides in all soil samples. The specific activity of individual radionuclides in soil samples is calculated by using equation 1.

3.1. Uranium (^{238}U and ^{235}U) Activity Concentration

The most samples were collected from Fuel Fabrication Facility (FFF) contain high levels of specific activity concentration of ^{235}U and ^{238}U, in current study most of soil samples contain high level of ^{234m}Pa which was the indicator of ^{238}U, by taking the single peak at 1001.03 keV gamma- ray transition for ^{234m}Pa. A well shielded and large (efficient) detector even allows the use of low intensity lines like the 1001.03 keV from the ^{234m}Pa decay. Because approx. 0.9987 of the ^{234m}Pa beta decay feeds the ground state of ^{234}U directly, the remaining beta branches, together with any gamma-rays and conversion lines are weak [13, 14]. The absence of a peak for the 766.6 keV gamma ray is explained by the fact that the intensity of the 1001.03 keV gamma ray peak is quite low (0.847%), but the yield for the 1001.03 keV gamma ray is more than two times the yield for the 766.6keV gamma ray (0.323%). Because of the long half life of ^{238}U and the short half-lives of ^{232}Th and ^{234m}Pa, both ^{232}Th and ^{234m}Pa can be assumed to be in decay equilibrium with ^{238}U. The measurements of activity in FFF are showedin table 2 that ^{238}U (^{234m}Pa) has a maximum concentration at a value of 375778±6895 $Bq.kg^{-1}$, and the lowest concentration is 177.74±60$Bq.kg^{-1}$. While the activity of ^{235}U has a maximum concentration at a value of 20954±407.5 $Bq.kg^{-1}$ and the lowest concentration is 7.4±0.5$Bq.kg^{-1}$.

Activity concentration limits were; 1Bq/g for each ^{238}U and ^{235}U for free released weights of materials.

Table 2. *Specific activity concentration, mass and activity ratio of 235U/238U.*

Code of sample	Activity ConcentrationBq/Kg					Mass Ratio × 10-3	Activity ratio U-235/ U-238 (234mPa) × 10-3
	U-238		U-235	Th-232 (228Ac)	K-40		
	Bi-214	Pa-234m					
S8	15.98±0.7	40332±702	1819±37	17±0.8	258±13	7.5	4.50
S12	16±1	174835±3269	2884±51	18±1.7	330±15	2.60	1.64
S14	13±1	84910±1690	1871±33	13±1	280±13	3.50	2.20
S15	14.64±1.34	20063.09±511	926.91±20	13.4±0.85	367.9±16.5	7.2	4.61
S16	16.78±0.3	7144.66±217	401.06±21.6	12.33±0.56	279.6±14.92	8.87	5.61
S17	18.21±0.7	8268.54±254.3	355.21±15.6	15.21±0.36	277.33±14.60	6.7	4.29
S18	17.54±1	375777.5±6895	20954±407.5	19.52±0.66	281.96±15.06	8.7	5.5
S19	14.84±1	140629.5±2670	3967.93±61	15.50±1.57	309±13.79	4.4	2.8
S20	18.72±1.15	111240±2121	3340.42±44	17.74±1.71	311.81±1.44	4.7	3.0
S31	20.51±0.48	441.06±61	20.55±1	10.22±1.02	207.39±13.53	7.3	4.6
S32	14.07±1	11573.6±332	658.21±12	14.61±0.33	332.31±15.79	8.9	5.68
S34	13.21±0.5	234077±4342	10697.31±258	15.94±0.56	47.5±5.3	7.1	4.5
S39	19.79±1	128676±2352	5700.34±2.5	20.45±0.33	402.2±15.5	6.9	4.42
S40	16.63±1	7842.1±233	363.08±7.81	14.12±0.91	242.79±12.81	7.2	4.62
S42	13.82±1	250.02±44	11.3±0.553	17.08±0.21	325.015±13.6	7.1	4.51
S47	14.41±1	193.57±37	7.4139±0.5	15.11±0.92	333.5±1.54	6.0	3.8
S49	12.64±1	108506±2096	3252±121	16.55±0.41	240.25±13.41	4.7	2.9
S50	15.11±0.63	2220.8±129	95.93±3	20.34±1.29	337.36±15.7	6.7	4.31
S51	20.11±0.52	76560±1585	3253.8±80	7.06±0.22	240.09±10.68	6.6	4.25
S54	18.23±1	177.74±60	9.27±1.2	12.55±0.83	299.22±13.00	8.1	5.21
S59	16.02±0.32	183.07±46	7.5±0.6	15.71±01.5	206.46±1.55	6.4	4.09

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3.2. Thorium (^{232}Th) Activity Concentration

The measurements of activity concentration in Fuel Fabrication Facility (FFF) site are showed that activity concentration of Thorium varied from (7.06±0.22) to (20.45±0.33) Bqkg-1. The specific activity adopted on the actinium isotope (228Ac) at energy (911.60) keV is equivalent to the specific activity of Thorium isotope (232Th).

3.3. Potassium (^{40}K) Activity Concentration

The measurements of radioactive elements activity in Fuel Fabrication Facility (FFF) site are showed that Potassium has a maximum concentration value is (402.2±15.5) Bq.kg^{-1}, while the lowest concentration value is (47.5±5.3) Bqkg^{-1}. From the same table it is clear thatwe calculated the mass ratio from equation (6), and the activity ratio (^{235}U / ^{238}U) to determine type of uranium (natural or depleted), the results of activity ratio showed that all measured values close to ~ 0.0461 (natural uranium) and only five surface soil samples indicating the low- level of depleted uranium in the measured samples S12, S14, S19, S20 and S49 as shown in table 2. The results of mass ratio indicated that all values were within natural uranium(^{235}U / ^{238}U= 0.0072) and only two values were close to depleted Uranium S12, S14. Also from the results, the presence of depleted uranium is found to be insignificant. The mean activity ratio certain was derived from ^{235}U and ^{238}U isotopes for all the samples. This is consistent with previous studies and indicates the presence of natural Uranium from this location. We can see from this study that there is no evidence of enriched Uranium because this site was used to the manufacture of fuel and not to enrich uranium. The selected samples were contained high concentrations of ^{238}U(^{234m}Pa) and ^{235}U as shown in Table 2.

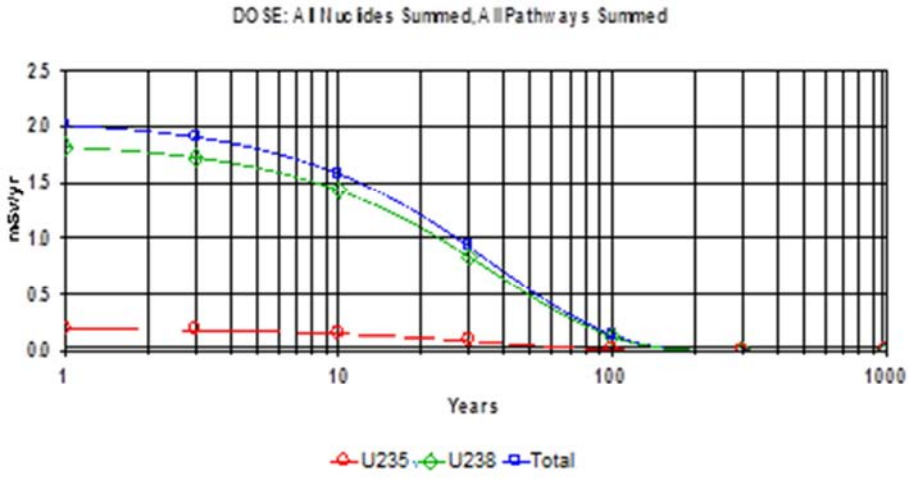
Depleted uranium is uranium that is a residual product obtained from the production of uranium fuel for nuclear reactors. Most reactors need Uranium with a higher concentration of^{235}Uthan found in natural Uranium.

4. Resrad for Radiological Risk Assessment

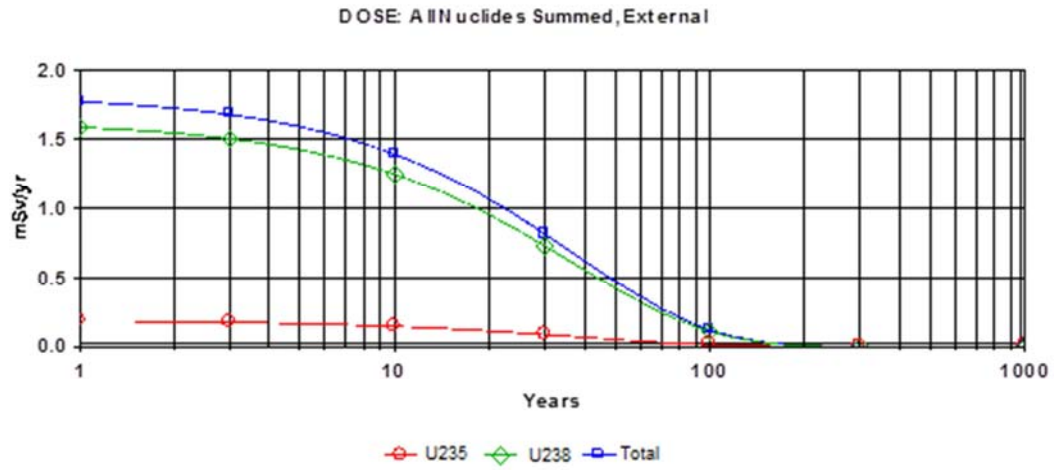
In resrad, the exposure times spent indoors and outdoors considered for the inhalation pathway are the same as those considered for the external exposure pathway. The indoor airborne dust level could be less than the outdoor level. The cover layer above the contaminated zone that is considered for reducing external radiation is also considered for reducing the inhalation exposure in resrad.

Outdoor Worker Scenario Comparison

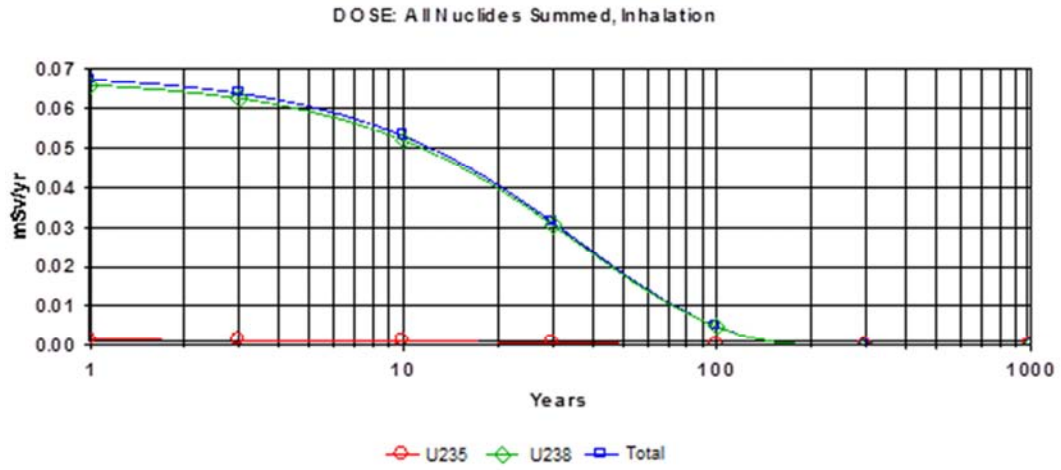
The Outdoor Worker Scenario considers three exposure pathways: (1) direct exposure to external radiation from the contaminated soil (2) internal radiation from inhalation of contaminated dust, and (3) internal radiation from incidental ingestion of soil. RESRAD (onsite) Version 7.0 was used to calculate the potential radiation dose for the outdoor worker. The period considered for this analysis was 1,000 years. The RESRAD input parameters used for this comparison are (Contaminated area 1250 m^2, Precipitation0.156 (for Baghdad, Indoor and outdoor time fraction were 0.8, 0.2 respectively). The RESRAD code provides individual exposure pathway dose contribution to the total dose. The individual pathway dose was estimated to be 2mSv/yr>0.3 mSv/yr based value which recommended by IAEA and represents the B. G for clearance case and it is clear that total dose decreases with increasing the numbers of years due to many conditions including erosion as result of (rains, wind and others conditions) and decreasing of half life of nuclide as shown in Figure (1a), 80% of the total dose arising from ^{238}U(^{234m}Pa) due to its high radioactivity and that is mean the remediation is required because the area which has taken within the hot spot. From Figure (1b) the external dose estimated and found 1.77 mSv/yr. The inhalation dose that resulting from external dose also estimated and found 0.068 mSv/yr as shown in Figure (1c). The soil ingestion dose can be seen from Figure (1d) estimated as nearly 0.168mSv/yr. The probability of cancer disease injury incoming for workers and the public, so total life time cancer risk has estimated and found 4.23×10^{-3} as shown in Figure (1e), this value is higher than the real value (4×10^{-6}) [15].



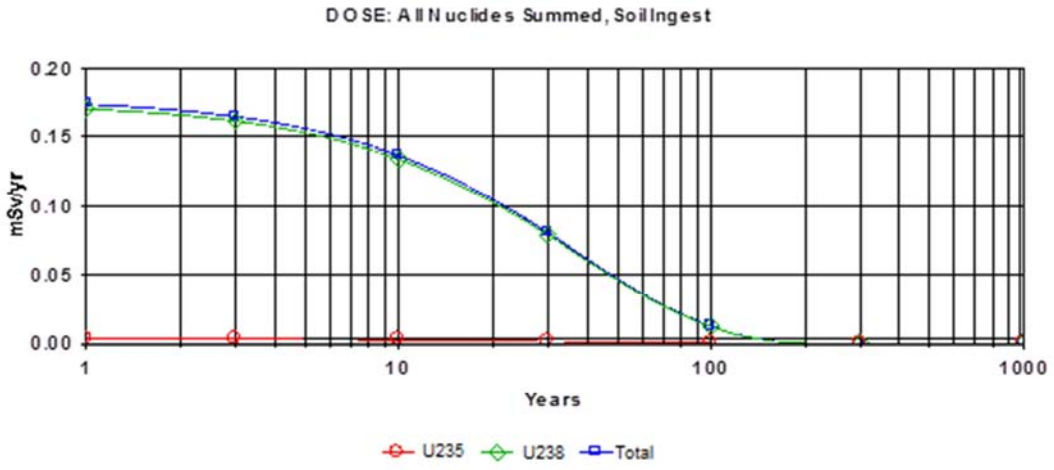
(a)


DOSE: All Nuclides Summed, External
mSv/yr
2.0
1.5
1.0
0.5
0.0
1
10
100
1000
Years
U235
U238
Total

(b)


DOSE: All Nuclides Summed, Inhalation
mSv/yr
0.07
0.06
0.05
0.04
0.03
0.02
0.01
0.00
1
10
100
1000
Years
U235
U238
Total

(c)


DOSE: All Nuclides Summed, Soil Ingest
mSv/yr
0.20
0.15
0.10
0.05
0.00
1
10
100
1000
Years
U235
U238
Total

(d)

(e)

Figure 1. *Time-integrated.*

(a) A pathways summed dose calculated in mSv/yr by RESRAD(onsite) 7.0 software.
(b) External dose (c) Inhalation dose (d) Ingestion dose (e) cancer risk

5. Conclusions

Al-Tuwaitha nuclear site considered as unique case, most of its facilities suffer substantial physical damage during the Gulf Wars and have been subjected to subsequent looting. The main objectives of this study are to assess the radioactive contamination in the soil Fuel Fabrication Facility at Al-Tuwaitha site, Iraq, the exposure and dose from the contaminated area, and the risk assessment. Based on the results obtained from this study, the following conclusions have been derived after processing the soil radioactivity.

1- From the calculation of the isotopic ratio $^{235}U/^{238}U$ to determine the type of Uranium in hot spots whether that was natural, depleted or enrichment. Mass ratio and activity ratio were used and turns out that very a few samples contain depleted uranium and others contain natural and there is no evidence for the presence of enriched uranium because this facility was used for fabricating nuclear fuel and not to enrich Uranium.

2- The calculated of total dose for all collected samples according to the applying of RESRAD on-site code one can observe that the value was 2mSv/yr > 0.3mSv/yr where as 0.3mSv/yr represents the basic or background dose in case of clearness. The cancer risk estimated and found 4.23×10^{-3} which is higher than recommended value by IAEA (4.0×10^{-6}). From all results, it is clear that this site must be subjected to regulatory control and safety surveillance to ensure adequate protection of the operators, public and the environment.

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