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## Total Radiological Dose and Safety Assessment of Workers in Radioactive Liquid Waste Storage Location in Al Tuwaitha Site

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### Abstract

This study aims to determine the total effective dose (external and internal) in the radioactive liquid Waste Hall (WH) in the Radiochemistry Laboratories building (RCL) in Al- Tuwaitha site in order to estimate the radiation risk for workers in this place. The external effective dose was calculated by an indirect method using ThermoLuminescent Dosimeters (LiF: Mg, Ti (TLD-100)) by placing them in the waste hall for one month. The internal effective dose was estimated by collecting eight air samples (environmental samples) from different locations in WH using an air sampler pump (Sniffer) to collect air samples loaded with particles or dust which are deposited on cellulose filter papers ( $\varphi$  125mm). This was done for two flow rates the lowest and the highest values of the air sampler pump of 5CFM and 20CFM, respectively. The filter papers were chemically treated using the digestion method. The radionuclides in these samples were measured using a gamma spectrometry system equipped with a high-purity germanium detector (HPGe), and the radioactivity concentration was measured using a gross alpha beta gamma system. The results showed <sup>137</sup>Cs of low concentration was deposited on the filter paper. The total annual effective dose rate (external and internal) results showed that the highest in WH was (6358.97 $\mu$ Sv/year) for the low probability scenario and (1589.74  $\mu$ Sv/year) for the realistic scenario.

**Keywords:** Al-Tuwaitha site, low probability and realistic scenario, TLD-100.

### تقدير الجرعة الكلية والسلامة الإشعاعية للعاملين في موقع خزن النفايات المشعة السائلة

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## الخلاصة

الهدف من البحث هو تقييم الجرعة الاشعاعية الكلية (الخارجية والداخلية) في بناية مختبرات الراديوكيمياء داخل قاعة النفايات المشعة السائلة في موقع التويثة النووي وذلك لمعرفة مدى الخطورة الاشعاعية على العاملين في هذا المكان اثناء عملهم. الجرعة الاشعاعية الخارجية قدرت بطريقة غير مباشرة وذلك باستخدام كواشف التألقي الحراري (LiF:Mg,Ti (TLD-100) بعد وضعها في هذه القاعة لمدة شهر واحد, بينما الجرعة الداخلية قدرت عن طريق جمع ثمان عينات هواء من عدة مواقع داخل وخارج قاعة النفايات باستخدام مضخة لجمع نماذج الهواء المحمل بدقائق او غبار المترسبة على أوراق ترشيح سيلولوزية ( $\phi$  125mm) والذي تم معاملته كيميائياً بطريقة الهضم. كررت هذه العملية لمعدلين جريان الهواء احدهما معدل جريان واطئ (5CFM) والآخر معدل جريان عالي (20 CFM) هذه النماذج تم تحليلها باستخدام منظومة تحليل اطياف كاما ذات كاشف الجرمانيوم عالي النقاوة لتحديد النويدات المشعة و تم قياس تراكيز هذه النويدات ( $\text{Bq}\cdot\text{m}^{-3}$ ) باستخدام منظومة عداد مجمل باعثات الفا وبيتا وكاما, النتائج لتحليل هذه النماذج بينت وجود نظير السيزيوم-137 بتراكيز واطئة. نتائج الجرعة الاشعاعية السنوية بينت اعلى قيمة ( $6358.97\mu\text{Sv}/\text{year}$ ) في حالة طريقة الاحتمالية الواطنة و ( $1589.74\mu\text{Sv}/\text{year}$ ) في حالة الطريقة الحقيقية.

## 1 Introduction

Workers in a variety of locations, such as nuclear power plants and medical clinics, are exposed to natural and man-made sources of radiation. The operation and decommissioning of nuclear power plants and other nuclear sites discharge radioactive elements into the environment and create radioactive waste [1-3]. Radiochemistry Laboratories (RCL) in Iraq were founded by SNIA techint-Italy in 1978 as part of the chemical research facility in the Iraqi Atomic Energy Commission in Al-Tuwaitha (approximately 20 kilometers south of Baghdad), which is now under the Iraqi decommissioning project plan. The RCL activities produced enormous quantities of radioactive liquid waste containing a variety of radionuclides [4, 5]. The wastes were collected in a radioactive liquid waste hall (WH) and categorized into three pools: low-level liquid waste (LLW), organic liquid waste (OW), and high-level liquid waste (HLW). The radioactive liquid waste in these pools contains the main radionuclide  $^{137}\text{Cs}$  [4-7].

In a radiological workplace, a radiological dose assessment of a non-sealed source must be carried out because the radioactive substance might enter the body of the worker and be absorbed by organs or tissues [8]. The potential of exposure (pathways) such as external direct radiation exposure or internal exposure (through inhalation or ingestion) should be recognized to determine the overall radiation dose and set scenarios of who are exposed to radiation, whether employees, the general public, or children. The total effective dose (TED) comes from both internal and external exposure, therefore internal and external dosimetry monitoring of workers should be performed to assess the suitability of TED for internal dosimetry [8, 9].

Radiological internal dose assessment relies on the calculation of the intake of a radionuclide, which can be estimated either from direct measurements, e.g. external monitoring of the whole-body counter system of specific organs and tissues, indirect measurements, e.g., through analysis of urine or faeces samples, or from environmental samples (air sample) measurements [10, 11]. The last method was adopted in this research. The effective dose can be estimated from internal exposure (through inhalation or ingestion) using dose coefficients for radionuclides that are recommended by the International Commission on Radiological Protection (ICRP) and International Atomic Energy Agency and (IAEA). The airborne particulate samples are usually collected on filter paper with the aid of

an air pump and a flow meter. The air should flow through the filter paper at a known rate for a known time [12].

The external dose can be estimated by direct measurements using nuclear detectors such as portable radiation devices or by thermoluminescent dosimeters (TLDs) to measure the personal dose at a depth of 10 mm (Hp(10)) from the location of the workplace. The TLD method was adopted in this study. TLD detectors have many advantages, including ease of use, compact size, reusability, and a limited number of correction factors, in addition to their close tissue equivalency (effective atomic number of 8.2, which is equivalent to tissue's 7.4), minimal signal fading (5–10 % per year), wide linear response range (10µGy- 10Gy), and great sensitivity for extremely low dose assessments [13]. Lithium fluoride with magnesium and titanium (LiF: Mg, Ti) TLD is the most extensively used. There are many different varieties of TLD, including personal dosimeters, environmental monitors, space dosimeters, and clinical dosimeters. The dosimetry system of TLD consists of a dosimeter and its reader.

External dosimetry refers to the measurement of outside body exposure to radiation. For this sort of dosimetry, personal dosimeters are employed, which are worn by employees at all times while they are exposed to radiation to ensure that their dose limits are not exceeded [14].

In this study, occupational exposure for workers depended on using two mechanisms (scenarios): realistic and low-probability scenarios to estimate the total radiation dose [15]. The dose assessment of external and inhalation exposure in these scenarios for workers depends on many factors, such as exposure time (working hours), density of dust, and breathing rate [16], as shown in Table 1.

**Table 1:** The parameters of realistic and low probability scenarios to workers [8].

Parameter	Realistic Scenario	Low Probability Scenario	Unit
	Value	Value	
Exposure time	450	1800	hour/year
Density	1.5	1.5	g/cm <sup>3</sup>
Breathing Rate	1.2	1.2	m <sup>3</sup> /h
Dose coefficient <sup>e<sub>j,inh</sub></sup>	6.7x10 <sup>-9</sup>	6.7x10 <sup>-9</sup>	Sv/Bq

Direct external exposure, where the received dose is at (1m) height, and internal exposure via inhalation and ingestion were chosen as the paths of exposure to radiation that relate to workers in the radiation field. The annual total effective dose (E<sub>t</sub>) is calculated as [8,9]:

$$E_t = E_{ext} + E_{inh} + E_{ing} \dots\dots\dots(1)$$

where E<sub>ext</sub> is the external exposure dose of direct radiation, E<sub>inh</sub> is the dose caused by inhaling contaminated material, and (E<sub>ing</sub>) is the dose caused by ingesting contaminated material. The contribution of both external and internal exposure must be considered when calculating the effective dose (E<sub>t</sub>), which was calculated using the following expression [15, 16]:

$$E_t = H_p(d) + \sum_i e_{j,inh} \cdot I_{j,inh} \dots\dots\dots(2)$$

where: H<sub>p</sub>(d) is the personal dosage equivalent at depth d in the body during period t.

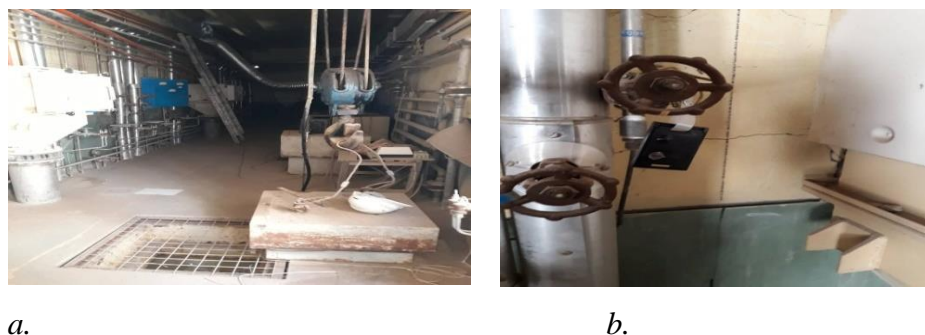
$e_{j,inh}$  is the dose coefficients (in units of  $\mu\text{Sv/Bq}$ ), which is the committed effective dose per unit activity intake by inhalation of radionuclide  $j$ , and  $I_{j,inh}$  is the intake of radionuclide  $j$  (in units of Bq) during the period.

The breathing rate for workers and other adults was set at  $1.2 \text{ m}^3/\text{h}$  and dose coefficients for workers were taken for  $5 \mu\text{m}$  activity median aerodynamic diameter (AMAD).

## 2 Material and Methods

### 2.1 External Radiological Dose Measurement

Ten thermoluminescent dosimeters (TLD-100) were placed in three locations near each pool in the waste hall (WH) in the Radiochemistry Laboratories (RCL) at 1 m height for one month, as shown in Figure 1. In this study, the TLDs used ((LiF:Mg,Ti (TLD-100)) were with nominal dimensions of  $(3.2 \times 3.2 \times 0.89) \text{ mm}^3$  and a lower dosage limit of  $10 \text{ pGy}$ , contain two chips (detectors): one for deep dose, Hp(10), and the other for shallow dose, Hp(0.07). An automated reader (Harshaw-6600 lite) with an optional  $^{90}\text{Sr}/^{90}\text{Y}$  internal irradiator was used to read the TLDs, and a hot nitrogen gas as a non-contact heating medium was used to heat the TLDs.



**Figure 1:** a. RCL waste hall, b. TLD dosimeter

### 2.2 Internal Radiological Dose Measurement

#### 2.2.1 Samples Collecting

An air pump (RADeCO, Inc) with Whatman cellulose filter paper ( $\phi 125 \text{ mm}$ ) manufactured from high-quality cotton liners was used to collect a known volume of air samples. Eight air samples were collected at a one-meter height for one hour; six were collected inside the WH near each liquid pool with different air flow rates of the air pump (cubic feet per minute, CFM). The samples were coded depending on their location and the flow rate of the pump, such as HLW-5 and HLW-H, where the first three letters refer to the name of the pool and the last refers to the air flow rate (5 CFM and High, which is equal to 20 CFM). The other two air samples were collected outside the WH as a background to compare the air radioactivity in the WH with different air flow rates, which were coded as BG-5 and BG-H (5 and H are the air flow rates, 5 CFM and H for high flow rate, which was equal to 20 CFM). A low-volume air sampler pump (Sniffer) was used for the measurement of the total suspended solid concentrations of each pool.

The volume of air sample ( $\text{m}^3$ ) was calculated as:

$$V = CFM \times T \times 0.0283 \quad \dots \dots (3)$$

where:  $V$  is the air volume (in  $\text{m}^3$ ) entering the filter paper,  $CFM$  is the air flow rate (cubic feet per minute),  $T$  is the time of collecting the sample (60 minutes), and 0.0283 is the conversion factor from cubic foot to cubic meter

### 2.2.2 Samples Preparing

The air samples (the cellulose filter papers) were prepared by the chemical digestion process by adding 15 mL of nitric acid and 10 mL of hydrochloric acid to each filter in a 250 mL beaker and heated on a hot plate at 150°C for two hours [17]. The solution was filtered with filter paper after adding distilled water, and the volume of the filtered solution was reduced by heating using the hot plate. The residual solution was deposited on a stainless steel planchet (of 2-inch diameter) to be ready for measurement in the gross alpha beta gamma system, as shown in Figure 2.



a. TLDs



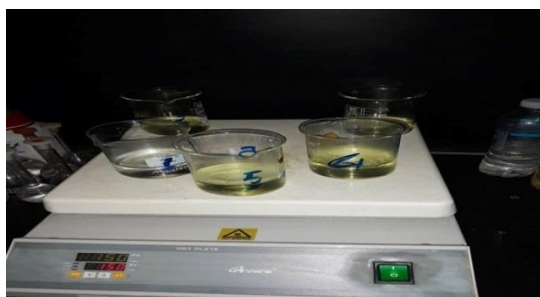
b. air pump



c. air samples ( filter paper)



d. preparing samples



e. hot plate



f. planchets

**Figure 2:** a. The TLD dosimeter, b. the air pump sampler, c, d, and e. samples preparations, and f. the deposited samples on stainless steel planchet

### 2.3 Gamma Spectrometry System

A gamma spectrometry system with a high purity germanium detector (ORTEC) of 65% relative efficiency, provided with a software program (Gamma Vision-32 software version-7),



was used to determine the radionuclides in the air samples, as presented in Figure 3-a. Its resolution is ( $\leq 1.9$ ) keV at an energy line of 1.33 MeV of  $^{60}\text{Co}$  and 1 keV at the energy line of 122 keV of  $^{57}\text{Co}$ , and  $^{137}\text{Cs}$  was determined from the 661.6 keV gamma line peak [18,19].

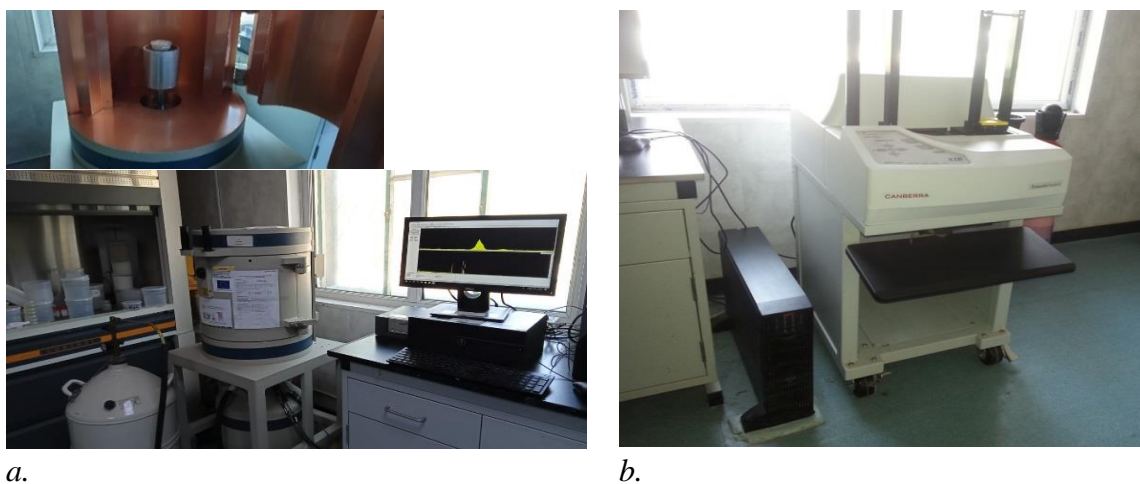
#### 2.4 Gross Alpha Beta Gamma System (Eclipse LB)

The gross alpha beta gamma system (manufactured by Canberra Company), shown in Figure 3-b, consists of two detectors, the first is a proportional gas-flow type for the detection of alpha and beta rays; a 2.25 inches' diameter pancake style gas-flow proportional detector with P-10 gas, with efficiency of 38% for beta and 40% for alpha. The second detector is sodium iodide scintillation type for detecting gamma rays. This system displays total or gross alpha beta and gamma count rate (counts per minute, cpm), gross activity (Bq, Ci,  $\mu\text{Ci}$ ), activity concentration ( $\text{Bq.g}^{-1}$ ,  $\text{Bq.L}^{-1}$ ) or disintegration per minute (DPM) without determining the radionuclide.

The gross beta activity concentration is calculated using the equation below [20]:

$$A(\text{Bq.m}^{-3}) = \frac{(C - B)}{\text{eff} \times V \times 60} \quad \dots (4)$$

where:  $C$  is the beta count rate (counts per minute) and  $B$  is the background count rate,  $\text{eff}$  is the beta efficiency and  $V$  is the volume of sample ( $\text{m}^3$ ).



**Figure 3:** a. Gamma spectrometry system, b. Gross alpha beta gamma system

### 3 Results and Discussion

#### 3.1 External radiological dose

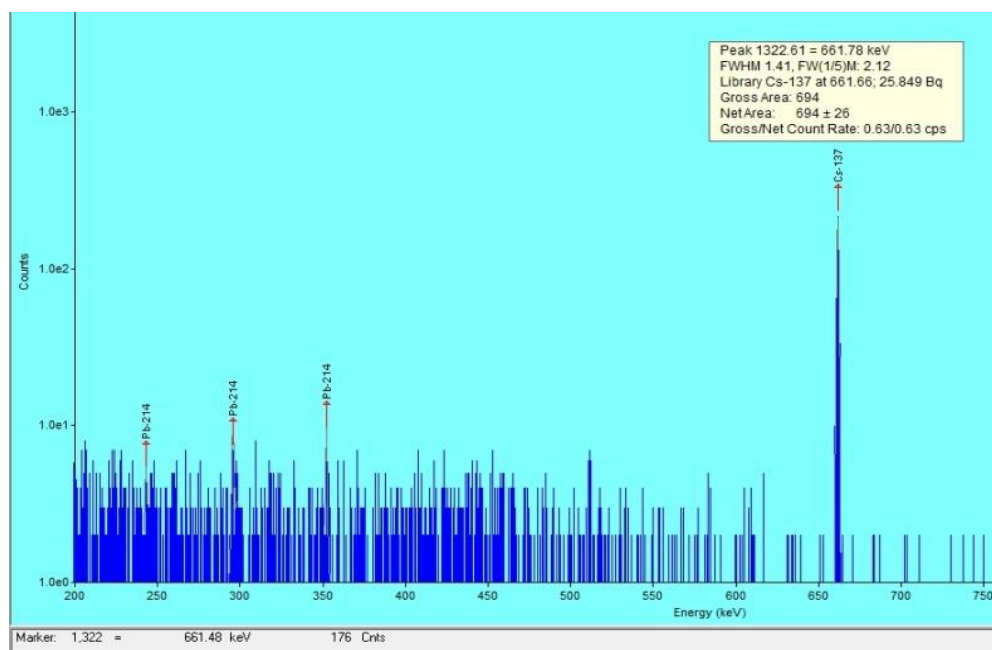
The results of the ten thermoluminescent dosimeters (TLD-100) used to monitor the external radiological dose for one month in the waste hall, measured by the automatic reader (Harshaw model 6600 lite), are shown in Table 2. These results were converted to the realistic and low probability scenario for a year exposure time (450 and 1800 h, respectively) as shown in Table 1. The highest dose was in the middle and the end of the WH near the third group of tanks (HLW), because of the highest radioactivity concentration of liquid waste in the third pool.

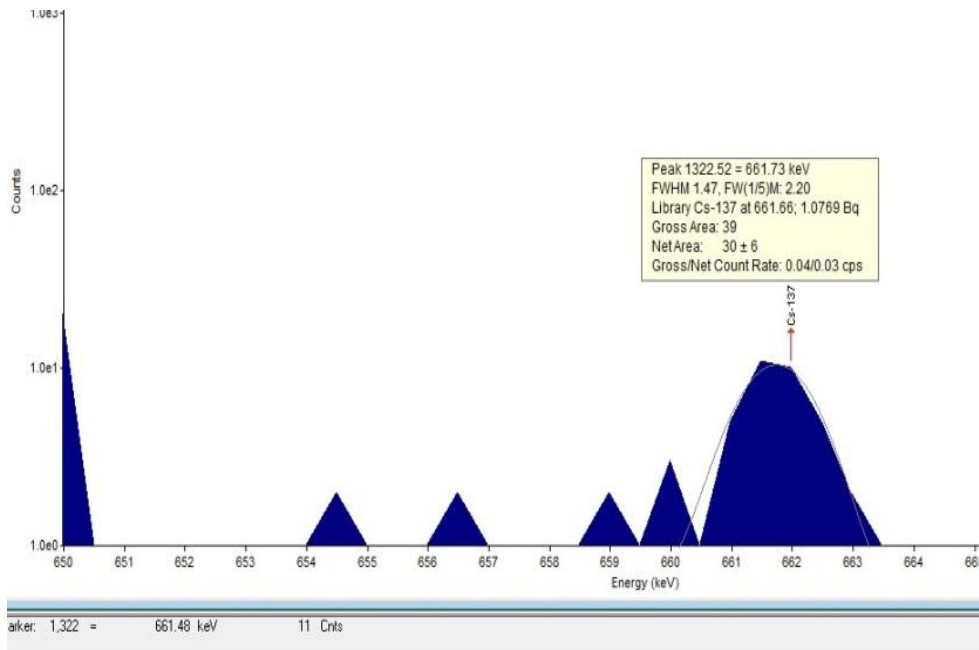
**Table 2:** External dose measurements inside the WH.

Location	deep dose Hp (10) ( $\mu\text{Sv}/\text{month}$ )	shallow dose Hp (0.07) ( $\mu\text{Sv}/\text{month}$ )	the total dose ( $\mu\text{Sv}/\text{month}$ )	Average dose ( $\mu\text{Sv}/\text{month}$ )	Dose for low probability scenario ( $\mu\text{Sv}/\text{year}$ )	Dose for realistic scenario ( $\mu\text{Sv}/\text{year}$ )
TLD-BG	85	65	150			
TLD-1	284	253	537			
TLD-2	33	177	210			
TLD-3	182	374	556	Ave. = 434.33	1085.83	271.45
TLD-4	474	300	774			
TLD-5	139	42	181			
TLD-6	149	418	567	Ave.= 507.33	1268.33	317.08
TLD-7	491	635	1126			
TLD-8	820	4384	5204			
TLD-9	610	1160	1770	Ave.= 2521.75	6304.37	1576.09
TLD-10	282	1705	1987			

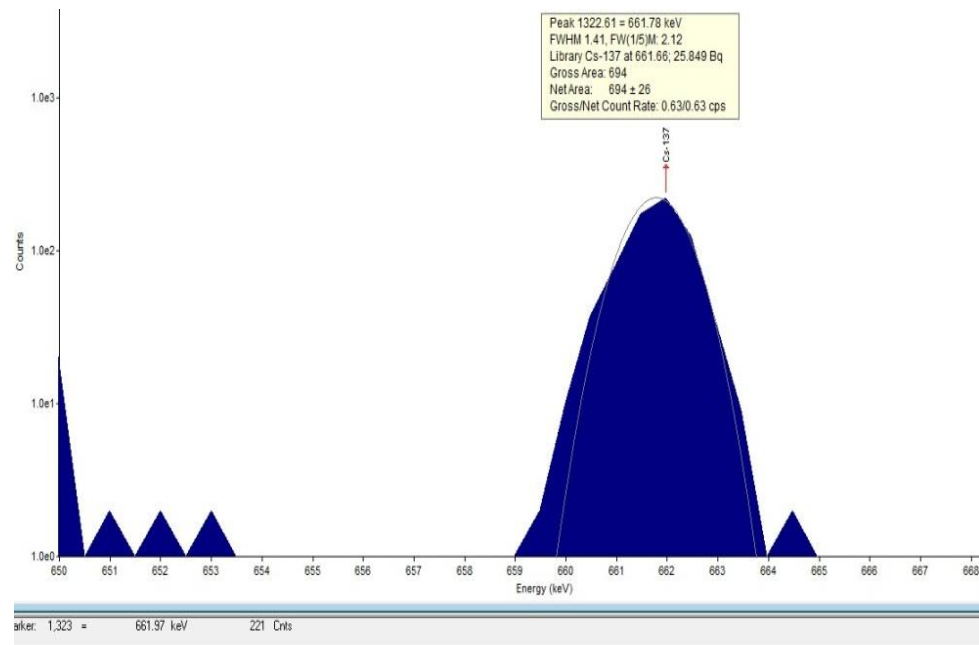
### 3.2 Internal Radiological Dose

Two techniques were used to study the air samples (filter papers) of known air volume prepared by a chemical digestion process. The first technique was the gamma spectroscopy system which determined the radionuclides in the samples without their concentration because the air volume standard source was unavailable. The filter samples were put directly on the HPGe detector in the gamma spectrometry system. Figure (4) shows the spectrum of the HLW-5 air sample, while Figures 5 and 6 show a comparison between the gamma spectrum of caesium-137 in the background air filter paper samples at a flow rate of 5 CFM (BG-5) with that of the air sample near the high level liquid waste pool (HLW-5).

**Figure 4:** Gamma spectrum of HLW-F-5 sample



**Figure 5:** Cs-137 peak of gamma spectrum to Background-5 sample.



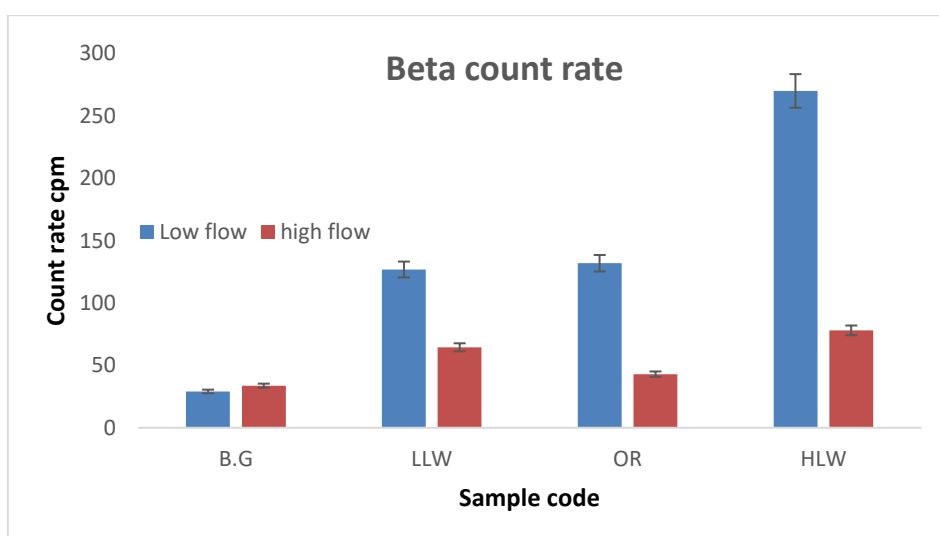
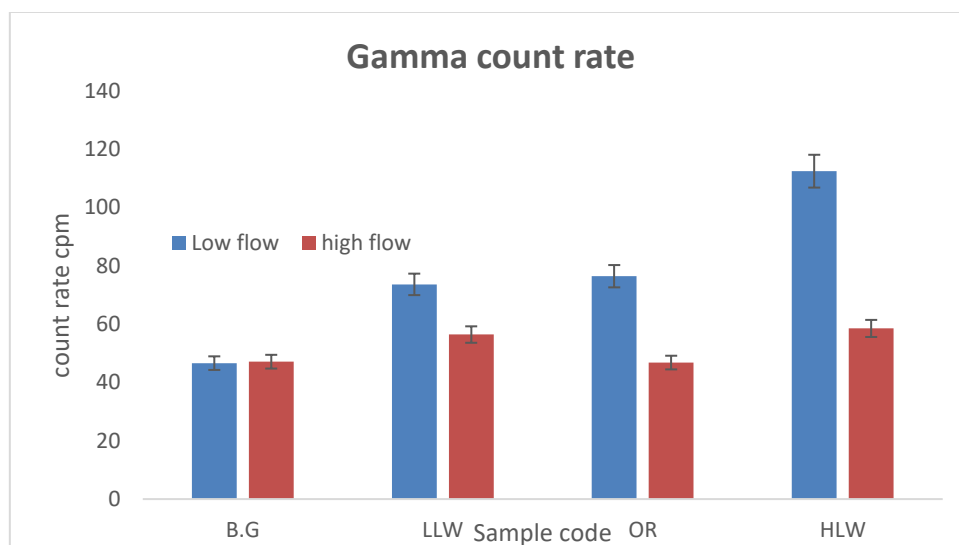
**Figure 6:** Cs-137 peak of gamma spectrum of HLW -5 sample.

The second technique used the gross alpha beta system, which was carried out for the samples of the two flow rates, 5 CFM and high flow (H CFM), near each pool. The 5 CFM is the lowest flow rate of the air pump, and the high flow was set at 20 CFM. This system does not determine the type of radionuclides; it just detects the radioactivity and gives the gross alpha, beta, and gamma count rate or their concentrations, as presented in Table (3) and Figures (7 and 8).



**Table 3:** Alpha count rate, Beta count rare and gamma count rate in air samples measured by gross alpha beta gamma system in different flow rates (F)

Sample code	Alpha (cpm)	Beta $\beta$ (cpm)	Net beta count rate (C- BG)	Gamma (cpm)
<b>B.G of device</b>	0.04	20.00	---	45.42
<b>BG-5</b>	0.00±0.0	29.1±1.71	9.1	46.6±2.16
<b>BG-H</b>	0.4±0.2	33.7±1.84	13.7	47.1±2.17
<b>LLW- 5</b>	0.3±0.17	126.9±3.56	106.9	73.6±2.71
<b>OR-5</b>	0.2±0.14	131.9±3.63	111.9	76.4±2.76
<b>HLW-5</b>	0.10±0.10	270.00±5.2	250	112.4±3.35
<b>LLW-H</b>	0.5±0.22	64.5±2.54	44.5	56.40±2.37
<b>OR-H</b>	0.00±0.0	43.00±2.07	23	46.8±2.16
<b>HLW-H</b>	0.5±0.22	78.1±2.79	58.1	58.5±2.42

**Figure 7:** Gross beta count rate of air samples**Figure 8:** Gross gamma count rate of air samples

The net beta activity concentration for cesium-137 radionuclide (in the Becquerel/m<sup>3</sup> unit) was calculated using Eq. 4 from the count rate (given in Table 3). The air volumes (in Eq. 4) for the two values of the flow rate was calculated using Eq. 3, which were 8.49m<sup>3</sup> and 33.96m<sup>3</sup> for 5CFM and H (20CFM), respectively. Table 5 presents the results of the total inhalation radioactivity (intake) concentration (Bq/m<sup>3</sup>) and total activity (Bq) after applying the result of beta radioactivity.

Table 5 also presents the results of the annual internal effective dose for low and realistic probability scenarios using the scenario parameters (exposure time) from Table 1 and Eq. 2 using internal conversion coefficient equal to 0.0076  $\mu$ Sv/Bq.

**Table 5:** The total air samples radioactivity and annual internal effective dose for low and realistic probability scenario.

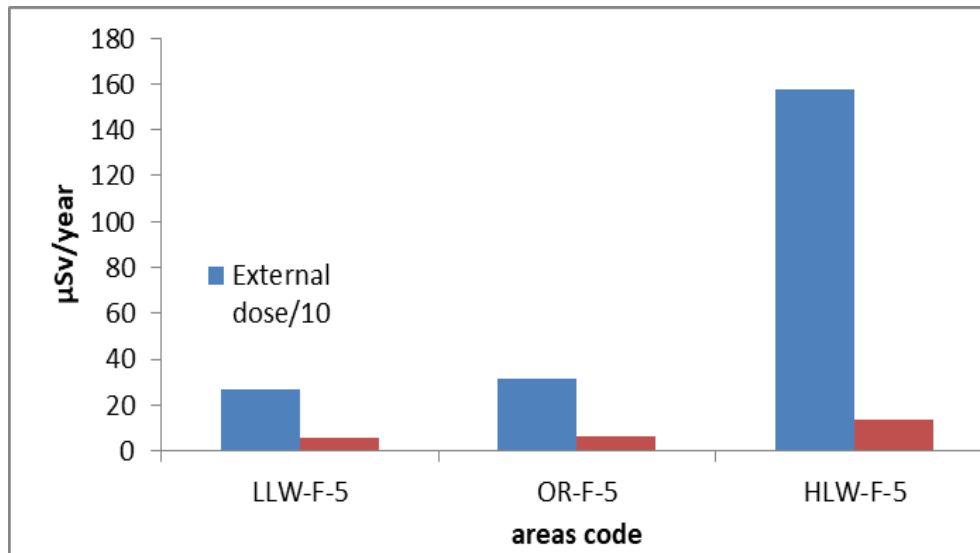
Code	Net beta count rate	Activity con. A (Bq/m <sup>3</sup> )	Intake =A.V (Bq) for Low Probability Scenario	Intake =A.V (Bq) for Realistic Scenario	Annual dose for Low Probability Scenario ( $\mu$ Sv/year)	Annual dose for Realistic Scenario ( $\mu$ Sv/year)
LLW-5	106.9	1.613	3484.75	871.19	23.35	5.84
OR-5	111.9	1.689	3647.74	911.93	24.44	6.11
HLW-5	250	3.773	8149.55	2037.39	54.60	13.65
LLW-H	44.5	0.168	362.65	90.66	2.43	0.61
OR-H	23	0.087	187.44	46.86	1.26	0.31
HLW-H	58.1	0.219	473.49	118.37	3.17	0.79

### 3.3 The total radiological dose

Table (6) presents the external and internal effective dose rates(calculated using Eq. 2) and the total effective dose rate which is the sum of the external effective and the internal effective dose, (as calculated by Eq. 1) for low and realistic probability scenarios. These were calculated for the low flow rate of the air pump(5CFM) only , since it is near the breathing rate of the workers. Figure (9) compares the external and internal effective dose rate for the realistic scenatio. From this figure, it can be noted that the external dose is greater than the internal.

**Table 6:**The annual total effective dose for low and realistic probability scenarios

Area in WH	External effective dose rate ( $\mu$ Sv/year)		Internal effective dose rate ( $\mu$ Sv/year)		Total effective dose rate ( $\mu$ Sv/year)	
	Low scenario	Realistic scenario	Low scenario	Realistic scenario	Low scenario	Realistic scenario
LLW-5	1085.83	271.45	23.35	5.84	1109.18	277.29
OR- 5	1268.33	317.08	24.44	6.11	1292.77	323.19
HLW-5	6304.37	1576.09	54.6	13.65	6358.97	1589.74



**Figure 9:** The annual effective dose rates for a realistic scenario inside WH

#### 4. Conclusions

The beta particles (from caesium-137) count rate of low airflow samples was greater than that of high air flow samples. The low airflow was at 5 CFM, the lowest scale of the air pump, equal to 8.49 m<sup>3</sup> air volume near the breathing rate of workers. Therefore, the 5 CFM represents a more significant of the radioactivity intake by the inhalation pathway.

The most effective contribution to the total effective dose of each pathway for the three locations inside the waste hall was due to external exposure.

Despite the high annual total effective dose in the WH, it is still below the dose limit (20mSv/year) recommended by the IAEA and ICRP.

It is advisable to use the ALARA principle (As Low As Reasonably Achievable) in this case due to the large amount of the liquid waste in the WH. The radiation dose of the workers can be reduced by decreasing their working hours and wearing protective clothing. The internal dose must also be monitored by wearing suitable respiratory devices or masks and prohibiting eating, drinking, and smoking inside the WH.

#### 4. Acknowledgment

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