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Determination of Activity Concentrations and Internal Dose Assessments in Urine from Workers in Iraqi Nuclear Research Center, Iraq

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Abstract:

To detect tritium levels in the human body, tritium in the urine must be measured. The investigation of the level of tritium activity in the urine of workers of the Iraqi Nuclear Research Center (INRC) has been realized in order to assess vocational exposure levels. Three worker groups in INRCs were chosen, involved in the Central Laboratories Directorate (CLD), the Decommissioning Directorate (DD), and the Radioactive Waste Treatment and Radioactive Waste Store Directorate (WTWSD), among others in Al-Tuwiatha site, in addition to the control group, as the research targets. Tritium concentration was determined in urine samples from 52 workers (aged 32-61) and 13 control people (aged 30-58). 65 urine samples of 50 mL were collected and analyzed. The Eichrom's Tritium Column way was used to handle the samples of urine, and the distillate tritium activity concentration was then assessed using Liquid Scintillation Counter. The effective and annual effective doses were estimated for the internal doses. The accuracy of urine sample measurements had been tested. The highest level of tritium activity samples of urine from three groups of workers in INRCs is 65.401 Bq/L, and for control group is 39.300 Bq/L, activity of tritium concentrations estimated by using counting scintillation of liquid (LSC) were found to be lower than Minimum Detectable Activity (MDA) of 1.95 Bq/L. The effective and annual effective dose from tritium was also assessed depend on results measured previously and standard values adapted by the International Commission on Radiological Protection. The effective highest doses for workers groups and control group were 497.048 $\times 10^{-3}$ and 298.680$\times 10^{-3}$ nSv respectively. Highest annual effective dose 5757.773$\times 10^{-3}$ and 5167.042$\times 10^{-3}$

nSv respectively. These results were lowest than the allowed effective dose for the workers.

Keywords: tritium, urine**,** activity concentration, effective dose, annual effective dose

1. Introduction

Tritium (^{3}H) is radioactive hydrogen isotope, emitting a low-energy beta particle (electrons) with a highest energy of 18.6 keV, a mean energy of 5.7 keV, and a radiological half-life of 12.3 years. It is present in nature in very low level [1]. It can be found in nature as a result of cosmic ray interaction in atmospheric gases or as a result of spontaneous fission of natural uranium in the Earth's crust, atmospheric nuclear and recent weapon testing. It is mostly made by human activities like reactors of heavy water and nuclear fusion technologies [2, 3]. The atmospheric amount of tritium elevated considerably due to nuclear weapons tests. Nuclear weapons experiments yielded levels of tritium in the atmosphere more than 1000 TU. Modern-day values have decrease to 50 - 100 TU levels, with the decrease due to the eradication of atmospheric nuclear weapons testing and radioactive decay [4, 5]. Tritium due to being hydrogen isotope has properties similar to conventional hydrogen. Since tritium emitted low-energy beta particle, distance of most of them range in air only about 5 mm or in water or soft tissue 0.005 mm [4].

Due to this low range it is non-dangerous in vitro. Tritium is considered to have a mild effect in vivo. Tritium enters the human body initially by inhalation of tritiated vapors in the air during work in a volatile atmosphere the second way through skin absorption. Inside the body tritium quickly and homogenously distributed in whole body. The minimum radiation level related to these tissues is basically homogenous and related to the tissues' hydrophobicity [4]. Also uptaked of tritium from the human body through food and water ingestion [3]. It is mostly known that tritium is a non-toxic isotope, but chronic exposure to a tritium source may lead to seriously harm human health. As a result, urine test of nuclear power plant workers should perform on a regular basis for radiation protection [6]. With the active revolution production of nuclear power and the wide use of nuclear technique, more workers in nuclear power plants (NPPs) are exposed to radioactivity. Due to occupational risk factor, radioactivity can cause internal and external hazardous and subsequently cause immediate or late radiation insult [7, 8]. The tritium low decay energy limits its toxicity; thus, tritium has a harm effect from internal sources only. Despite its low energy, chronic

exposure to tritium radiation has been found to have a greater biological efficacy than that of ɣ -rays and X-rays [9]. Moreover, beta radiation from tritium has been shown to be more effective when combined within the molecule [10]. The aim of the study to investigate the activity of tritium concentration in urine of the Iraqi Nuclear Research Center (INRC) workers, in order to evaluate the occupational exposure levels.

2. Materials and Method

2.1 Al-Tuwaitha Nuclear Site Description

Al-Tuwaitha Center of Nuclear Research (ATNRC) was started in about 1960. It was located 18 kilometers southeast of Baghdad, the capital of Iraq, at 33° 12.57' north and 44° 31.822' east. Its coverage area of about 1.3 km2. Location was near Tigris River eastly about 1 km, to south of Baghdad about 20 km. This place belonged to the prior Iraqi Commission of Atomic Energy (IAEC); around it there was earthen berms that are approximately 4.6 km long and 30 m high and contain three gaps that allow for vehicle access. Prior to 1991, The Al Tuwaitha compound consisted of 90 infrastructures related to the radiochemistry lab, production of nuclear fuel, enrichment of uranium, handling and storage of radioactive waste, and biological research. In 1991 and during the Gulf War, most of the facilities in Al-Tuwaitha were extensively destroyed. Immediately following the Gulf War of 2003, there was a breakdown in security in Iraq, which adversely affected the security of access control to the Al-Tuwaitha site and other facilities throughout Iraq. As a result, there was huge looting of materials like equipment, some amount radioactive contamination occurred, and there was damage associated with many facilities. Information on distribution of radionuclide and radiation concentration in the environment is pivotal for detecting the terrestrial radiation exposure effects due to cosmogenic and human activities [11,12]. In this study, three important facilities were chosen in INRC: Central Laboratories Directorate (CLD), Decommissioning Directorate (DD), and Radioactive Waste Treatment and Radioactive Waste Store Directorate (WTWSD).

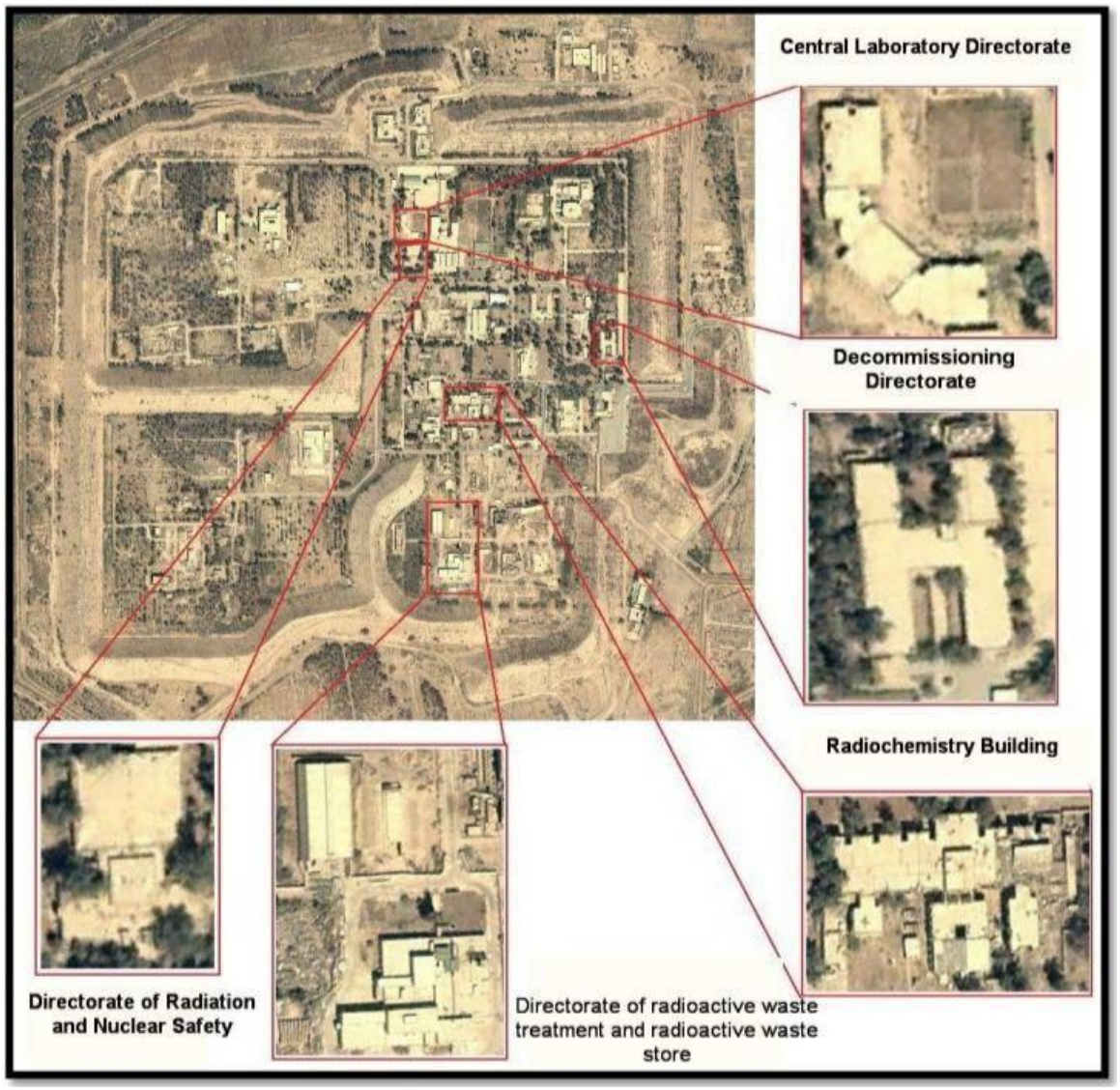

Figure 1: Iraqi Nuclear Research Center (INRC) in Al-Tuwaitha site

2.2 Collection of Urine Samples

In June 2022, 65 participants provided a 50-mL urine sample. This participant group was divided into two groups: the workers group, which included 52 participants who work daily in the field of radiation at the Iraqi Nuclear Research Center (INRC) and ranged in age from 32 to 61 years, from three directorates: 25 workers from the radioactive waste treatment and management directorate (WT), aged 33–61 years; 15 workers from the decommissioning directorate (WD), aged 35–59 years; and 12 workers from the Central Laboratories Directorate (WL), aged 32–59 years. In addition, the control groups included 13 apparently healthy volunteers, randomly chosen from the population living in Baghdad, far away from the Al-Tuwaitha Nuclear Research Site. The age range of the unexposed control group was 30–58 years old. The samples were then transferred to separate 50-mL plastic bottles. Workers and control volunteers were asked to complete a questionnaire with information on their gender, age, working hours, and the types of ailments they suffer from, particularly disorders of the urinary system. They were also asked about the donor's drinking water source and if he or she is a smoker or alcoholic.

2.3 Pretreatment of Urine Samples

Urine samples were processed using Eichrom's Tritium Column Method. Tritium columns (^{3}H columns) are used for the splitting and detection of free tritium are substitute for direct assessment or measurement after distillation [13]. It has been developed to shorten the preparation time and decrease waste generation for a variety of sample matrices, and this method is used to substitute distillation for much of usual tritium analyses of aqueous samples. The practical procedure that was followed in the current work is to take approximately 25 ml of urine sample, add it to 4 gm of activated charcoal, introduce a suitable magnetic stirring bar in the container, shake well for approximately 15 minutes by Hotplate Stirrer Model L-81 (Labinco), Netherlands (to adsorb organic compounds in urine), and then filter to obtain a clear filtration that removes color and not all contaminate chemical material (these contaminates will act with the liquid of scintillation and cause luminescence). Then the filtered sample is collected in a 22-ml conical glass flask, and the sample is transferred to tritium separation columns to be separated by the vacuum system (coulombs, vacuum box, and vacuum pump) at a pressure of 10 mBar. The collected sample after separation is placed in a cold place before adding to the scintillation liquid Ultima Gold. To eliminate

luminescence, samples should be saved in scintillation vials for 1 day in a dark place or canister before measurement.

2.4 Preparation of Urine Samples

10 mL of the residue suppuration sample was added to 10 mL of Ultima Gold LLT cocktail (PerkinElmer) in a glass measuring vial (200 PCS standard size, 20 mL for the liquid scintillation counter), for a total volume of 20 mL. The vials were shacked for around one minute to make the solutions homogeneous. In added, the vials were cleaning by ethanol before being placed on the table to prevent contamination. The background sample was prepared using twice-distilled ground well water with a low level of tritium [14]. Measurement of tritium concentration was made using counter of a small-level of scintillation liquid (Perkin Elmer Tricarb 3110 TR, USA). It is a computerized analyzer of liquid scintillation for exploring low level of alpha, beta, and gamma radiation. The samples are counted for 210 minutes (3 h 30 m).

2.5 Quality Control

The instrument was checked through performing a self-normalizing calibration (SNC) to ensure that the instrument accurately quantifies the energy for all beta particles emission, and the performance of instrument assessment (IPA) procedure using a non-quenched standard calibration of tritium, calibration standard of an unquenched carbon 14, and a basic standard supplied by PerkinElmer. The standard solution of tritium each with specific activity of 262900 DPM (0.2 µCi) were peaked into two samples handled for distillation of urine.

3. Measurement and Calculations

3.1 Counting Efficiency (IPA instrument performance evaluation)

The efficiency of the instrument used for counting of tritium E was measured by using the following equation:

$$E = \frac{Nd - Nb}{D} \qquad (1)$$

Where, Nd is the measurement of tritium sample basic rate (counts per min); Nb is the rate of counting background sample (counts in min); D is the activity of tritium addition to the basal sample (decays in min).

3.2 Tritium Activity Concentration

The level of tritium activity concentration (A) in the sample of urine can be calculated as below [15]:

$$A(\frac{Bq}{L}) = \frac{Ns - Nb}{60 \times E \times V} \quad (2)$$

Where, Ns is the activity of the sample (counts/min), and V is the volume of the urine distillate sample (L).

3.3 Minimum Detectable Activity

The Minimum Detection Activity (MDA) was calculated according to the Currie formula [16]:

$$MDA = \frac{3.29\sqrt{\frac{Rb}{ts}+\frac{Rb}{tb}+\frac{2.71}{ts}}}{60EV} \quad (3)$$

Where Rb is the count rate of the background (counts/min), ts is the counting time of the sample (min), tb is the counting time of the background (min).

3.4 Inner Dose Calculation

The internal active dose (ED) can be written in terms of tritium activity concentration and default half-life. The half-life is supposed to be 10 days when continuous deposition occurs in the absence of other evidence.

The formula for this estimation can be written as follows [17]:

$$ED\ (Sv) = \frac{5.3\times10^{-11}}{\ln 2/10} \times A = 7.6 \times 10^{-10} \frac{Sv}{Bq/L} \times A \quad (4)$$

Where 5.3×10^{-11} Sv per day per (Bq/L) is the dose coefficient of tritium.

3.5 Annual Effective Dose

The annual effective dose AED was formulated by the ICRP 78 as given by the following equation [18]:

$$AED\ (Sv) = 0.73 \times m \times EDC \times A \quad (5)$$

Where the 0.73 is the coefficient of the fraction of water mass in the body, m is the body weight (kg), EDC is the effective dose coefficient (1.8×10^{-11} Sv Bq^{-1}).

4. Results and Discussion

The results in Table 1 showed the highest urine tritium level of three groups of workers in the INRCs, namely the Radioactive Waste Treatment and Radioactive Waste Store Directorate (WTWSD), the Decommissioning Directorate (DD), and the Central Laboratories Directorate (CLD), were 65.401, 3.351, and 39.323 Bq/L, respectively, while the highest concentration of tritium for the control group is 39.300 Bq/L. The highest internal effective dose due to tritium ingestion for the three worker groups was 497.048×10^{-3}, 25.468×10^{-3}, and 298.855×10^{-3} nSv, respectively, while in the control group it was 298.680×10^{-3} nSv. The annual effective dose of tritium was also assessed depend on the standard of measurement results and reference values recommended by the International Commission on Radiological Protection (ICRP) [19]. The highest annual effective dose for the three worker groups was 5757.773×10^{-3}, 356.660×10^{-3}, and 5167.042×10^{-3} nSv, respectively, while the highest annual effective dose for the control group was 3356.613×10^{-3} nSv. These levels are lower than the allowed tritium dose inside the body, that is, 40000 nSv [19].

Although all the results were less than the permissible value of tritium dose 40000 nSv recommended by ICRP, 1994 [19]. However, it has been noted that there are abnormal results in each of the WT, WL, and C groups. This negatively affected the results of the standard deviation calculations, although all the results have been recalculated to ensure the validity of the results. So we disclosed Table2 to announce the effect of the presence or absence of the results of abnormal samples on the standard deviation. Where the reasons for the abnormality of these results were analyzed by examining and interrogating the practical history of the worker with the abnormal result from his peers in the same group, and they were as follows:

Table 1: Tritium activity in urine and annual effective doses for the exposed workers groups in INRC and the control group.

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No.	Code	Age (Year)	Body weight (kg)	period of work (year)	CPM	DPM	Eff.%	Activity (Bq/L)	ED×10^{-3} (nSv)	AED×10^{-3} (nSv)
1	WT1	60	93	21	14	102	13.82	1.707	12.973	208.599
2	WT2	33	112	9	10	43	8.65	1.921	14.600	282.710
3	WT3	51	80	20	11	38	28.92	0.630	4.788	66.226
4	WT4	50	67	23	776	3924	19.77	65.401	497.048	5757.773
5	WT5	35	95	10	21	89	23.25	1.480	11.248	184.748
6	WT6	56	68	29	16	53	27.22	0.872	6.6272	77.915
7	WT7	56	85	20	23	89	25.52	1.480	11.248	165.301
8	WT8	49	84	21	11	85	13.14	1.411	10.723	155.741
9	WT9	46	93	28	14	64	22.04	1.065	8.094	130.145
10	WT10	42	50	22	25	103	24.55	1.722	13.087	113.135
11	WT11	39	82	10	39	202	19.39	3.365	25.574	362.572
12	WT12	34	80	23	6	38	16.89	0.641	4.872	67.382
13	WT13	54	75	22	49	182	27.00	3.038	23.089	299.395
14	WT14	52	90	22	9	49	19.14	0.824	6.262	97.446
15	WT15	41	75	18	10	45	22.63	0.753	5.723	74.208
16	WT16	51	100	21	15	52	28.34	0.866	6.582	113.792
17	WT17	45	81	10	9	40	22.52	0.666	5.062	70.885
18	WT18	57	92	30	7	43	16.91	0.710	5.396	85.830
19	WT19	52	80	29	14	61	22.86	1.019	7.744	107.117
20	WT20	50	94	24	13	42	31.54	0.704	5.350	86.955
21	WT21	47	85	29	10	53	18.28	0.887	6.741	99.069
22	WT22	48	86	21	12	66	18.38	1.106	8.406	124.982
23	WT23	61	91	30	19	64	29.68	1.074	8.1624	128.422
24	WT24	53	80	29	39	154	24.25	2.515	19.114	264.377
25	WT25	52	85	28	14	44	29.26	0.686	5.214	76.619
26	WD1	56	90	20	10	33	29.14	0.566	4.302	66.935
27	WD2	46	89	20	14	83	16.62	1.380	10.488	161.385
28	WD3	50	75	29	18	99	18.35	1.643	12.487	161.918
29	WD4	59	100	33	16	147	10.91	2.455	18.658	322.587
30	WD5	59	65	30	12	45	25.61	0.757	5.753	64.655
31	WD6	52	66	20	11	37	31.24	0.685	5.206	59.406
32	WD7	45	60	27	11	39	29.07	0.646	4.910	50.931
33	WD8	48	72	10	12	45	27.08	0.746	5.670	70.578
34	WD9	50	90	20	10	43	22.85	0.713	5.419	84.319
35	WD10	55	100	16	11	39	29.01	0.650	4.940	85.410
36	WD11	46	78	28	15	84	17.73	1.491	11.332	152.816
37	WD12	54	67	29	13	82	15.51	1.279	9.720	112.601
38	WD13	42	98	20	11	35	30.42	0.588	4.469	75.718
39	WD14	48	79	21	25	140	17.49	2.336	17.754	242.491
40	WD15	35	81	12	44	190	20.20	3.351	25.468	356.660
41	WL1	38	80	13	25	92	27.32	1.528	11.613	160.623
42	WL2	54	73	20	38	157	24.27	2.624	19.942	251.699
43	WL3	43	72	13	32	127	25.32	2.125	16.150	201.042
44	WL4	51	88	21	13	150	20.75	2.506	19.046	289.774
45	WL5	52	78	20	40	157	25.35	2.619	19.904	268.427
46	WL6	32	65	10	12	40	30.08	0.757	5.753	64.655
47	WL7	51	75	25	49	196	25.19	3.261	24.784	321.372
48	WL8	43	72	22	290	1169	24.84	19.490	148.124	1843.910
49	WL9	59	100	35	557	2359	23.32	39.323	298.855	5167.042
50	WL10	50	89	27	15	43	25.22	0.731	5.556	85.488
51	WL11	53	90	25	22	90	23.11	1.582	12.023	187.087
52	WL12	35	82	21	12	45	10.77	2.111	16.044	227.456
53	C1	35	65	10	14	102	13.82	1.707	12.973	145.795
54	C2	38	79	12	20	70	26.35	1.157	8.793	120.104
55	C3	44	90	20	11	51	21.79	0.857	6.513	101.349
56	C4	58	84	25	16	63	25.64	1.048	7.965	115.674
57	C5	44	70	33	8	30	27.44	0.505	3.838	46.450
58	C6	45	65	20	550	2355	23.32	39.300	298.680	3356.613
59	C7	28	50	—	46	206	22.45	3.433	26.091	225.548
60	C8	30	48	—	19	80	23.32	1.325	10.070	83.570
61	C9	39	55	25	13	49	27.40	0.815	6.194	58.900
62	C10	48	80	10	15	50	29.84	0.835	6.346	87.775
63	C11	50	75	25	13	54	24.99	0.968	7.357	95.396
64	C12	55	80	20	12	47	24.44	0.788	5.989	82.835
65	C13	48	90	—	12	42	29.04	0.697	5.297	82.427

WT: Workers in West Treatment and Management Directors.
WD: Workers in Decommissioning Directorate.
WL: Workers in Directorate of Central Laboratories.

The worker WT4 sample had activity 65.401 Bq/L , effective dose 497.048 nSv, and annual effective dose 5757.773 nSv, respectively. The workers WT4 and WT9 deal directly with radiation through the removal of radioactive waste, but they do not abide by the conditions of occupational radiation safety because they believe that their long-term work in the field of radiation gives them protection. The Radiation and Nuclear Safety Directorate has been notified of the exposed workers, to apply its preventive measures with them to protect them from the dangers of radiation. The Worker WL8 sample had activity 39.323 Bq/L, effective dose 298.855 nSv, and annual effective dose 5167.042 nSv, respectively. The Worker C6 sample had activity 39.300 Bq/L, effective dose 298.68 nSv, and annual effective dose 3356.613 nSv, respectively. The workers of the WL and C groups were fully committed to all safety conditions. However, there appeared in each group a high concentration of the sample of worker WL8 and worker C6 from their peers. It is believed that this increase in tritium concentration is due to environmental conditions, especially that through dialogue with worker WL8; it became clear that he is the brother of worker C6, and this confirms that the cause of exposure is environmental, not occupational.As for group WD, its results do not contain outliers.

It is noted from Table 1 that when comparing the results of exposure of the worker group to tritium with the control group, the results were very close, and if we compared the concentrations of tritium in the urine of workers in the radiation field at INRC, they was very close to the concentration of tritium in the control group and less than the global results attached in Table 3. Figure 1 shows the variation between the highest, lowest, and average values of the annual effective doses for each group of workers within the work period. This is evident depending on each of them on the period of work. All results are within internationally permissible limits.

Table 2: Statistical comparision of activity, effective dose and annual effective dose of the studying groups.

Range, Average, and Standard Deviation	WT Included abnormal case			WT* Not Included abnormal case		
	Activity Bq/L	ED×10^{-3} (nSv)	AED×10^{-3} (nSv)	Activity Bq/L	ED×10^{-3} (nSv)	AED×10^{-3} (nSv)
Range	0.360-65.401	4.788-497.048	66.226-5757.773	0.630-3.365	4.788-25.574	66.226-362.572
Average	3.862	29.349	368.054	1.298	9.862	143.482
SD	12.842	95.629	1103.039	0.742	5.641	81.127
	WD Included abnormal case			**WD* Not Included abnormal case**		
Range	0.566-3.351	4.302-25.468	50.931-356.66	**No abnormal case**		
Average	1.286	9.772	137.894			
SD	0.845	6.203	94.179			
	WL Included abnormal cases			**WL* Not Included abnormal cases**		
Range	0.731-39.323	5.556-298.855	64.655-5167.042	0.731-3.261	5.556-24.784	64.655-321.372
Average	6.555	49.816	755.7145	1.984	15.082	205.762
SD	11.016	83.720	1406.300	0.787	5.983	79.823
	C Included abnormal case			**C* Not Included abnormal case**		
Range	0.505-39.300	3.838-298.68	46.45-3356.613	0.505-3.433	3.838-26.091	46.45-225.548
Average	4.110	31.239	354.034	1.178	8.952	103.819
SD	10.183	77.394	867.832	0.744	5.652	44.663

Table 3: Tritium concentration in urine for the exposed workers and the general public according to reported in the literature

Country	Tritium concentration Bq L^{-1}	References
Italy	62.0 ± 4	[20]
Poland	12.0±4	[21]
Japan	0.8±0.3	[22]
Canada	11.2±3	[23]
Finland	2.6±2	[24]
South Korea	2.8±1.4	[25]
China	3.5±1.5	[26]
Turkey	4.7±1.9	[14]
Sweden	2.6±1	[27]
Iraq	0.12±0.1	Present study

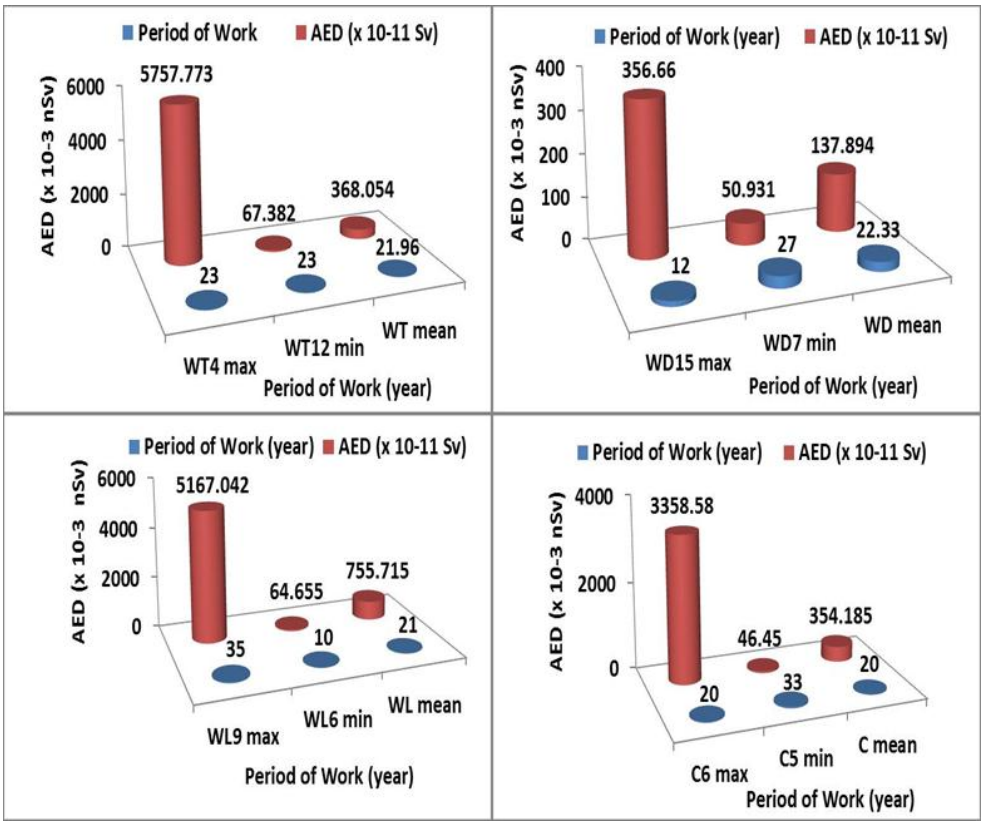

Figure 1: Annual effective doses with originate from tritium as a function of work period for the Iraqi radiation workers in INRC

5. Conclusions

Urine samples were obtained from two groups (the general population and employment exposed workers) and analyzed to find out the exposure of workers in the Iraqi Nuclear Research Center to radiation. Increased concentration of tritium was not present in the urine of workers or the general public. Only two cases were tritium activity levels found to be lower than MDA. Thus, all cases of contamination detect were occupational. Its levels in this study are very low in comparison to other exposed workers, such as those working in heavy water reactors. All results were within the internationally permissible limits, and this is due to the workers' high commitment to occupational safety measures.

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