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Measurement of Tritium Activity Concentrations in Water Samples of Al-Amara City in Misan Province-Iraq, using Liquid Scintillation Counter

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Abstract: This research work aims to measure the concentration of tritium activity in the Tigris river and samples of tap water collected from different places of Al-Amara city in Misan province using a Tri-Carb 3110TR liquid scintillation counter (LSC) and to calculate an annual effective dose due to the ingestion of water samples. The mean values of the tritium activity concentrations measured in the water samples were 0.562± 0.126 Bq/L (4.763 ± 1.075TU) and 0.521 ± 0.060 Bq/L (4.422 ± 0.512TU) for Tigris River and tap water samples, respectively. The mean annual effective doses are given to the infants, children and adults because of the intake of the tritium were (252.95 ± 57.116) $\times 10^{-5}$ µSv/y and (234.81 ± 27.222) $\times 10^{-5}$ µSv/y, (354.13 ± 79.962) $\times 10^{-5}$ µSv/y and (328.734 ± 38.111) $\times 10^{-5}$ µSv/y, and (738.614 ± 166.778) $\times 10^{-5}$ µSv/y and (685.645 ± 79.489) $\times 10^{-5}$ µSv/y for Tigris river and tap water samples, respectively. The results indicated that the concentrations of tritium activity in measured water samples have been found below the limit of 100 Bq/L, which is the recommended measure by the European Commission for drinking water, and the recommended annual effective dose by the World Health Organization for the members of the public was much less than the individual dose criterion of 100 µSv/y. Hence, the waters of the Tigris river and tap in the study area (Al-Amara city) are not threatened to cause health risks because of tritium concentration.

Key words: Tritium, LSC, Tigris river water, tap water, annual effective dose, Al-Amara City.

Introduction

Tritium is a radioactive isotope of hydrogen with a mass approximately three times that of the usual isotope. It can also be explained as a low-energy beta emitter (E_{max} = 18.6 keV) having a physical decay half-life of 12.3 years (UNSCEAR, 2008). Its complete emission is absorbed by general materials like plastic sheets, glass, or metal, and cannot penetrate the topmost dead layer of human skin. The exposure presents a hazard if the element is ingested through water or food, inhaled or absorbed through the skin (CNSC, 2009). Tritium occurs naturally and also originates from anthropogenic sources. Naturally, tritium gets produced in the atmosphere via the interaction of cosmic radiation with atmospheric nitrogen (Madruga et al., 2009). Anthropogenic production has affected the natural tritium level by tests of nuclear weapons. Also, tritium is released into the atmosphere during weapons manufacturing, operating nuclear power plants and reprocessing the nuclear fuels

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(Pujol et al., 1999). Tritium shows similar physical and chemical properties as hydrogen and hence, reacts with oxygen to form radioactive water (tritiated water, HTO) molecules. Tritiated water enters the environment's hydrologic cycle as ordinary water. Due to tritium's ease of addition into the human body, it becomes essential to measure the tritium activity levels in water samples available for human consumption. Its concentration limit in drinking water has been established by European Directive (European Commission,1998). Nowadays, numerous studies from different geographies are being carried out (Borio et al., 2005; Forte et al., 2007; Gören et al., 2014; Karataşli et al., 2017; Maringer et al., 2004; Semerjian et al., 2020; Stamoulisa et al., 2011; Villa et al., 2004). There is a lack of such tritium activity concentration studies in drinking water in Al-Amara city. The main objectives of this study, are both to evaluate the tritium activity concentrations in drinking water samples (Tigris river and tap waters) of Al-Amara city by using the liquid scintillation counting system which is a commonly used technique to measure radionuclides emitting low energy beta activity and to estimate the annual effective doses for different age groups of members of the public to evaluate the radiological hazards due to the intake of tritium. The result of this study will provide a database line for Al-Amara city, which can help in future research.

Material and Methods

The Study Area

Al-Amara city is the centre of Misan province and is located in the southeastern part of Iraq on the banks of the Tigris river, as shown in Figure 1. It is Iraq's 17^{th} largest city, with a population of 550,000 as of 2014 which occupies 2862 km^2. The latitudes for this area fall between 31°48′.00′′N to 31° 54′.0′′N and longitudes between 47°6′.0′′E and 47°12′.0′′E. This city is a commercial, economical, geographical and cultural region.

Sample Collection and Preparation

In the current conducted research, a total of thirty-four drinking water samples were collected out of which nine are from Tigris river water (RW) and 25 tap water (TW) samples, which were sourced from various water supplies in Al-Amara city are listed in Tables 1 and 2. The water samples were stored in 1-litre capacity polyethylene containers and each one of the collected samples was given a unique code (Figures 2 and 3). The sample containers were then labelled and

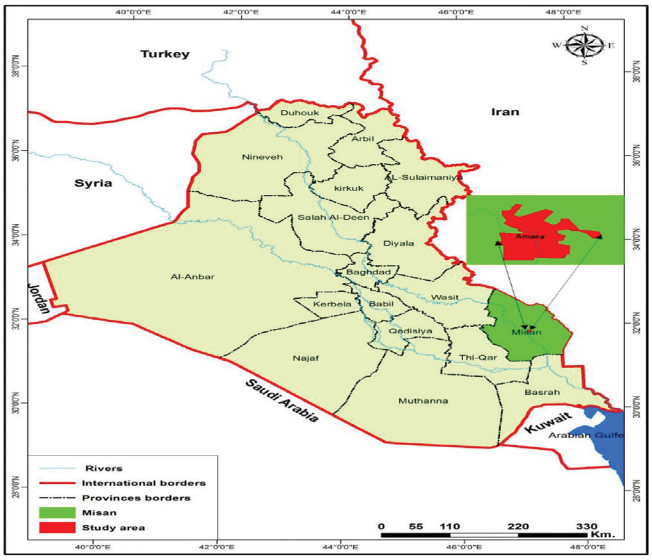

Figure 1: Map of the administrative divisions of Iraq showing the studied area (Al-Amara city).

Table 1: Positions of Tigris water samples in districts of Al-Amara city

Sample code	*Sample location*
RW_1	Al-Mualimin Al-Jadid
RW_2	Al-Karama
RW_3	Al-Saadiq
RW_4	Al-Sadr
RW_5	Al-Hassan Al-Easkary
RW_6	Al-Shabana
RW_7	Al-Askan
RW_8	Al-Qahira
RW_9	Dor Al-Naft

taken to the laboratory for tritium measurement. In the laboratory, these samples were prepared by using an ASTM method (ASTM D4107-08, 2006; Todorović et al., 2018). A total of 100 mL of the aliquot drinking water samples were treated with NaOH (0.5 g) because it excludes other radionuclides such as radioactive iodine and radiocarbon which will be distilled with tritium and $KMnO_4$ (0.1 g) used to oxidise organic substances that can intervene cooling. The distillate's middle fractions were extracted immediately for tritium analysis; the early and late fractions were discarded as they have interfering substances. The obtained distillate fractions were carefully blended with the addition of 10 mL of LS cocktail and prepared in the ratio of 8:12 (sample to cocktail). The mixture gets equilibrated by keeping it in the dark counting room for 6 hours and reaches room temperature. Further, beta emissions were counted using Tri-Carb 3110TR LSC.

Table 2: Positions of tap water samples in districts of Al-Amara city

Sample code	*Sample location*
TW_1	Al-Rahma
TW_2	Al-Mustafa
TW_3	Al-Jameiaat
TW_4	Al-Shuhadaa
TW_5	Al-Mualimin Al-Jadid
TW_6	Al-Zahraa
TW_7	Al-Ghadir
TW_8	Al-Karama
TW_9	Al-Saadiq
TW_{10}	Al- Jihad
TW_{11}	Al-Amam Al-Kazim
TW_{12}	Al-Sadr
TW_{13}	Al-Euruba
TW_{14}	El-Hussein
TW_{15}	Dor Al-Naft
TW_{16}	Al-Hassan Al-Easkary
TW_{17}	Al-Shabana
TW_{18}	Shaeban-15
TW_{19}	Al-Fatimia
TW_{20}	Al-Karar
TW_{21}	Al-Askan
TW_{22}	Al-Qahira
TW_{23}	Al-Mualimin Al-Qadim
TW_{24}	Al-Risala
TW_{25}	Al-Amir

Tri-Carb 3110TR LSC

The Tri-Carb 3110TR LCS, which is a low activity liquid scintillation analyser made by PerkinElmer, USA, was used to measure the amount of tritium activity in all of the samples collected (Tri-Carb 3110TR Instrument Manual, 2014). Most of the time, LSC is used to measure the concentration of tritium activity in aqueous samples. This is because LSC is sensitive, easy to use, has low detection limits, and is convenient even for measuring the concentration of natural tritium (Stojković et al., 2018). Around the vial chamber, active and passive shields make up the LSC's system for cutting down on background noise. The main component of a passive shield is lead, which reduces the background from gamma photons in the environment that come from building materials and instruments. A shield can also be lined on the inside with cadmium and copper, which absorb secondary X-rays and thermal neutrons. The active shield detector is made up of a shimmering material and two more photomultiplier tubes that surround the detector assembly. The detector takes the light from the blinking indicators and turns it into electrons in the photomultiplier tubes. The system has two pulse analysis circuits that are convenient to users: a pulse shape analysis (PSA) and a pulse amplitude comparator (PAC). The PSA can be used to find particles, including distinguishing between alpha and beta particles, while the PAC can be used to compare the ratio of the pulse amplitudes made by two PMTs (Passo et al., 1994). Chemiluminescence can be fixed with the help of a delayed coincidence circuit (DCOS). The

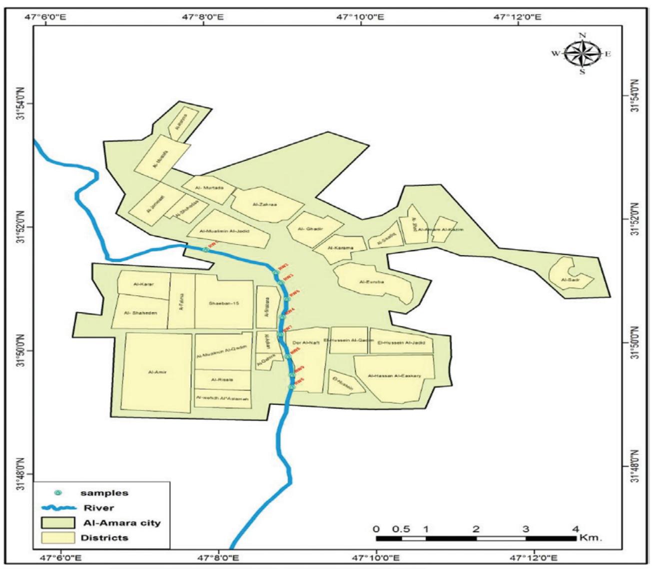

Figure 2: Map of the administrative divisions of the study area showing locations of Tigris river water samples.

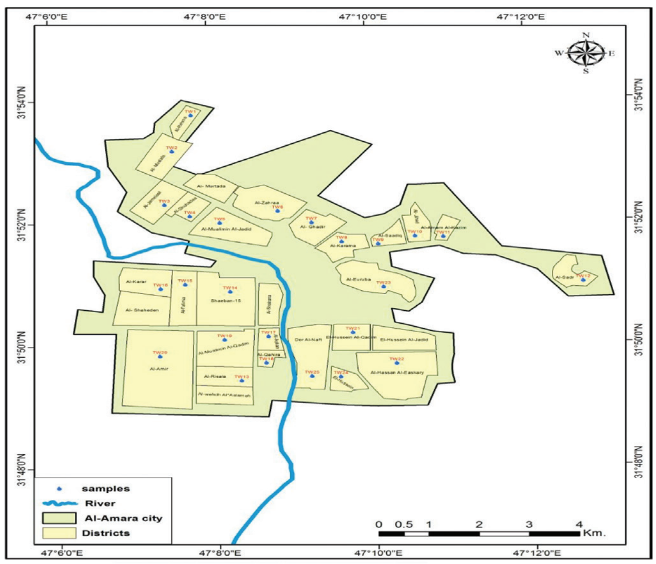

Figure 3: Map of the administrative divisions of the study area showing locations of tap water samples.

dual multichannel analyzers (MCA) used by LSC, first records active shields followed by recording spectra. MCA eliminates random noise produced by phototubes by using a tritium setting which helps to reduce the pulse from the guard and the sample that happens at the same time, simultaneously it keeps an eye on the DCOS-caused random coincidences in one half of the MCA and whole sample spectrum was recorded in the other half. The external standard quenching parameter SQP(E) is used to estimate the amount of quenching to figure out how well the system counts using the right calibration curves (Stojković et al., 2018).

Getting the LSC Right

For the LSC to be calibrated, the following samples had to be ready (Todorović et al., 2018):

i. Raw water tritium solution (RWTS) was made by adding tritium standard solution to not distilled raw water (RW).
ii. The raw water that has been distilled but does not contain tritium DRW for measuring the background.
iii. The distilled water tritium standard solution (DWTS) is made up of first distilled raw water followed by adding tritium standard solution, resulting in the same tritium activity via RWTS.
iv. Distilled raw water tritium standard (DRWTS): The RWTS has been treated with NaOH and $KMnO_4$, then distilled (first 10 mL of distillate was removed and the middle distillate was obtained).

Three aliquots from the DRWTS, DWTS, and DRW were made and the LS cocktail was mixed in the 20 mL polyethylene vials put in the dark and counted on the quantulus LSC. The count rates are found in the order: RDRWTS [s^{-1}], RDWTS [s^{-1}], and RDRW [s^{-1}].

Measurement of Tritium Activity Concentration

The minimum detectable activity (MDA)[Bq/ L] was determined using the equation given below (Todorović et al., 2018; Stojković et al., 2018) :

$$\text{MDA} = \frac{2.71 + 3.29\sqrt{R_{DRW} t_s \left(1 + \frac{t_s}{t_{\text{DRW}}}\right)}}{\varepsilon t_s \; FVe^{-\lambda t}} \quad (1)$$

where R_{DRW} [s^{-1}] was the count rates of the background sample; t_S [s] and t_{DRW} [s] denote the counting times of the sample and background respectively; ε was the detection efficiency; F was the recovery correction factor; V [l] is the sample volume and $\lambda(d^{-1}) = \frac{\ln 2}{t_{\frac{1}{2}}}$ counts for the decay constant for tritium.

The detection efficiency of counting was calculated as follows:

$$\varepsilon = \frac{R_{DWTS} - R_{DRW}}{A_{DWTS}} \quad (2)$$

where R_{DWTS} [s^{-1}] was the count rates of distilled water tritium standard solution and A_{DWTS} [Bq] was the activity of distilled water tritium standard solution.

The obtained recovery correction factor (F) was:

$$F = \frac{R_{DRWTS} - R_{DRW}}{\varepsilon A_{DWTS}} \quad (3)$$

where R_{DRWTS} [s^{-1}] was the distilled raw water tritium standard and A_{RWTS} [Bq] was the activity of raw water tritium solution.

The tritium activity concentration of the analysed sample, [Bq/ L], was calculated as:

$$A = \frac{R_S - R_{DRW}}{\varepsilon FVe^{-\lambda t}} \quad (4)$$

where R_S [s^{-1}] was the sample's aliquot gross count rate.

The tritium activity concentrations were turned into tritium units (TU) by using the following equation (Gören et al., 2014):

$$\text{A(TU)} = \frac{A(Bq/L)}{0.118} \quad (5)$$

Evaluation of Annual effective Dose

The annual effective dose estimation is done to evaluate the radiological risks to the public (infants, children, and adults) because of the consumption of the drinking water samples with the help of the formula (WHO, 2011):

$$\text{AED}(\mu\text{Sv/y}) = \text{A(Bq/L)} \times \text{CF (Sv/Bq)} \times \text{CR(L/y)} \times 10^6 \quad (6)$$

where A is denoted as the activity concentration of the tritium in (Bq/L); CF is the dose coefficient (1.8×10^{-11} Sv/Bq for tritium) and CR is the drinking water consumption rate (250, 350, and 730 L/y for infants, children, and adults, respectively) (Stamoulisa et al., 2011).

Results and Discussion

Tritium Activity Concentrations in Drinking Water of Al-Amara City

Using an equation, the minimum amount of activity that could be seen was found to be 0.137 Bq/L for a counting time of 1800 sec. Tables 3 and 4 show the amount of tritium activity (in Bq/L and TU) found in drinking water samples from Al-Amara, Iraq. These samples were taken from the Tigris river and from taps. The tritium activity concentrations measured in Tigris river water samples ranged from 0.347 ± 0.072 Bq/L (2.940 ± 0.610TU) to 0.781 ± 0.103 Bq/L (6.618 ± 0.872 TU) with a mean value of 0.562 ± 0.126 Bq/L (4.763 ± 1.075 TU), as seen in Table 3 and Figure 4. However, the tritium activity concentrations measured in tap water samples ranged from 0.388 ± 0.011 Bq/L (3.288 ± 0.093TU) to 0.643 ± 0.028 Bq/L (5.449 ± 0.237TU) with a mean value of 0.521 ± 0.060 Bq/L (4.422 ± 0.512 TU), as shown in Table 4 and Figure 5. The differences in tritium activity concentrations between Tigris river water and tap water samples are shown in Tables 3 and 4. Due to differing sampling points, the mean value of tritium activity concentrations recorded in Tigris river water samples is somewhat greater than the mean value of tritium activity concentrations measured in tap water samples. The tritium activity concentrations results showed that all drinking water samples were tested less than the recommended European Commission's limit of 100 Bq/L for drinking water (European Commission, 1998).

Table 3: Result of tritium activity concentrations of Tigris river water samples

Sample code	*Tritium activity concentrations*	
	(Bq/L)	*(TU)*
RW_1	0.347 ± 0.072	2.940 ± 0.610
RW_2	0.781 ± 0.103	6.618 ± 0.872
RW_3	0.609 ± 0.092	5.161 ± 0.779
RW_4	0.493 ± 0.021	4.177 ± 0.177
RW_5	0.561 ± 0.026	4.754 ± 0.220
RW_6	0.469 ± 0.021	3.974 ± 0.177
RW_7	0.538 ± 0.028	4.559 ± 0.237
RW_8	0.698 ± 0.031	5.915 ± 0.262
RW_9	0.563 ± 0.028	4.771 ± 0.237
Mean Value ± Standard Deviation	0.562 ± 0.126	4.763 ± 1.075
Minimum	0.347	2.940
Maximum	0.781	6.618

Comparison of Results

Table 5 compares the mean tritium activity content in the drinking water samples (Tigris river surface water samples and tap water samples) measured in the research area to similar measurements done in other countries. The mean concentration of tritium activity in the present studied water samples is lower than the previously reported mean concentration of tritium activity.

Table 4: Result of tritium activity concentrations of tap water samples

Sample code	*Tritium activity concentrations*	
	(Bq/L)	*(TU)*
TW_1	0.524 ± 0.022	4.440 ± 0.186
TW_2	0.465 ± 0.018	3.940 ± 0.152
TW_3	0.530 ± 0.027	4.491 ± 0.228
TW_4	0.546 ± 0.021	4.627 ± 0.177
TW_5	0.467 ± 0.013	3.957 ± 0.110
TW_6	0.556 ± 0.028	4.711 ± 0.237
TW_7	0.511 ± 0.026	4.330 ± 0.220
TW_8	0.496 ± 0.016	4.203 ± 0.135
TW_9	0.388 ± 0.011	3.288 ± 0.093
TW_{10}	0.517 ± 0.014	4.381 ± 0.118
TW_{11}	0.404 ± 0.011	3.423 ± 0.093
TW_{12}	0.509 ± 0.022	4.313 ± 0.186
TW_{13}	0.480 ± 0.018	4.067 ± 0.152
TW_{14}	0.566 ± 0.024	4.796 ± 0.203
TW_{15}	0.596 ± 0.027	5.050 ± 0.228
TW_{16}	0.494 ± 0.018	4.186 ± 0.152
TW_{17}	0.609 ± 0.029	5.161 ± 0.245
TW_{18}	0.570 ± 0.018	4.830 ± 0.152
TW_{19}	0.498 ± 0.013	4.220 ± 0.110
TW_{20}	0.564 ± 0.022	4.779 ± 0.186
TW_{21}	0.586 ± 0.021	4.966 ± 0.177
TW_{22}	0.643 ± 0.028	5.449 ± 0.237
TW_{23}	0.496 ± 0.014	4.203 ± 0.118
TW_{24}	0.566 ± 0.024	4.796 ± 0.203
TW_{25}	0.464 ± 0.015	3.932 ± 0.127
Mean value ± Standard deviation	0.521 ± 0.060	4.422 ± 0.512
Minimum	0.388	3.288
Maximum	0.643	5.449

*MDA is 0.137 Bq/L.
**MDA is 0.068 TU.

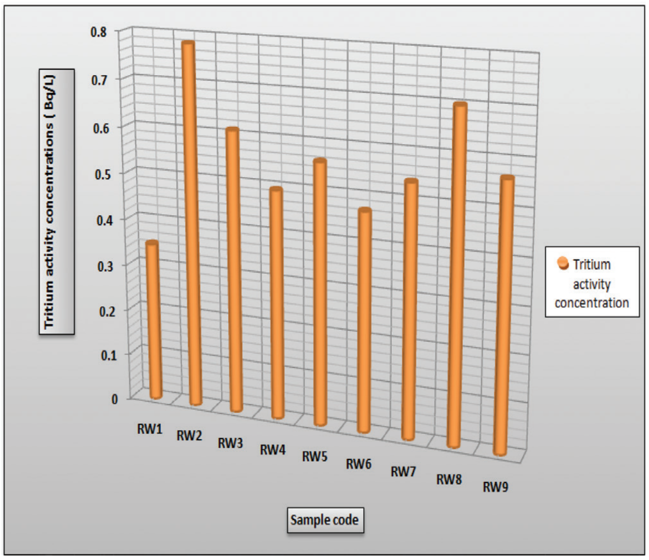

Figure 4: Tritium activity concentrations in Tigris river water samples.

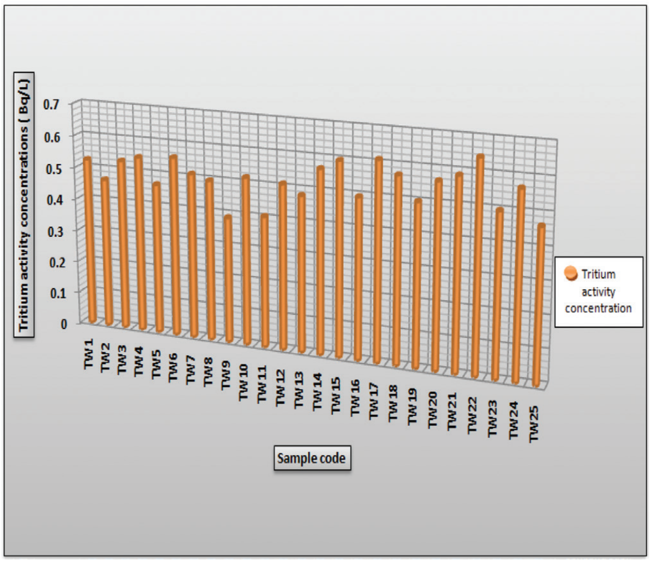

Figure 5: Tritium activity concentrations in tap water samples.

Calculation of Annual Effective Dose in Drinking Water Samples

Tables 6 and 7 showed the estimated annual effective dose (AED) values for the drinking water samples (Tigris river water and tap water samples). The estimated values of Tigris river water samples ranged from 156.15×10^{-5} to 351.45×10^{-5} µSv/y with a mean value of $252.95 \pm 57.116 \times 10^{-5}$ µSv/y, 218.61×10^{-5} to 492.03×10^{-5} µSv/y with a mean value of $354.13 \pm 79.962\times10^{-5}$ µSv/y, and 455.958×10^{-5} to 1026.234×10^{-5} µSv/y with a mean value of $738.614 \pm 166.778 \times10^{-5}$ µSv/y for infants, children, and adults, respectively, while in Tap water samples, estimated value ranged from 174.6×10^{-5} to 289.35×10^{-5} µSv/y with a mean value of $234.81 \pm 27.222 \times 10^{-5}$ µSv/y, 244.44×10^{-5} to 405.09×10^{-5} µSv/y with a mean value of $328.734 \pm 38.111 \times 10^{-5}$ µSv/y, and 509.832×10^{-5} to 844.902×10^{-5} µSv/y with a mean value of $685.645 \pm 79.489 \times 10^{-5}$ µSv/y for infants, children, and adults, respectively, as seen in Table 7 and Figure 7. Results of the estimated annual effective dose values depict that the examined drinking water samples fall under the dose criterion of 100 µSv/y recommended by the World Health Organization for all individuals (Onugba et al., 2009).

Table 5: Comparison of mean tritium activity concentrations (Bq/L) of water samples collected in present study with the previous data literature

Country and place	*Type of water*	*Tritium activity concentrations (Bq/L)*	*Reference*
Bulgaria	Danube river water	2	(Villa et al., 2004)
Italy (Umbria)	Drinking water	<8.6	(Borio et al., 2005)
Nigeria (Yola)	Ground water	0.58	(Onugba et al., 2009)
Greece	Rivers water	0.94	(Stamoulis et al., 2011)
Turkey (Adana)	Tap water	7	(Gören et al., 2014)
Turkey (Mersin)	Natural water	6.2	(Karataşli et al., 2017)
Turkey	Natural water	2.23	(Dizman et al., 2020)
	Mineral water	2.51	
Azerbaijan	Natural water	2.69	
	Mineral water	2.43	
Iraq (Al-Amara)	Tigris river water	0.562	This study
	Tap water	0.521	

Table 6: Result of annual effective dose estimated for members of the public in Tigris river water samples

Sample code	*Annual effective dose (10^{-5}) (μSv/y)*		
	Infant	*Child*	*Adult*
RW_1	156.15	218.61	455.958
RW_2	351.45	492.03	1026.234
RW_3	274.05	383.67	800.226
RW_4	221.85	310.59	647.802
RW_5	252.45	353.43	737.154
RW_6	211.05	295.47	616.266
RW_7	242.1	338.94	706.932
RW_8	314.1	439.74	917.172
RW_9	253.35	354.69	739.782
Mean Value ± Standard Deviation	252.95 ± 57.116	354.13 ± 79.962	738.614 ± 166.778
Minimum	156.15	218.61	455.958
Maximum	351.45	492.03	1026.234

Table 7: Result of annual effective dose estimated for members of the public in tap water samples

Sample code	*Annual effective dose (× 10^{-5}) (μSv/y)*		
	Infant	*Child*	*Adult*
TW_1	235.8	330.12	688.536
TW_2	209.25	292.95	611.01
TW_3	238.5	333.9	696.42
TW_4	245.7	343.98	717.444
TW_5	210.15	294.21	613.638
TW_6	250.2	350.28	730.584
TW_7	229.95	321.93	671.454
TW_8	223.2	312.48	651.744
TW_9	174.6	244.44	509.832
TW_{10}	232.65	325.71	679.338
TW_{11}	181.8	254.52	530.856
TW_{12}	229.05	320.67	668.826
TW_{13}	216	302.4	630.72
TW_{14}	254.7	356.58	743.724
TW_{15}	268.2	375.48	783.144
TW_{16}	222.3	311.22	649.116
TW_{17}	274.05	383.67	800.226
TW_{18}	256.5	359.1	748.98
TW_{19}	224.1	313.74	654.372
TW_{20}	253.8	355.32	741.096
TW_{21}	263.7	369.18	770.004
TW_{22}	289.35	405.09	844.902
TW_{23}	223.2	312.48	651.744
TW_{24}	254.7	356.58	743.724
TW_{25}	208.8	292.32	609.696
Mean Value ± Standard Deviation	234.81 ± 27.222	328.734± 38.111	685.645 ± 79.489
Minimum	174.6	244.44	509.832
Maximum	289.35	405.09	844.902

(*Contd.*)

*MDA is 0.137 Bq/L.

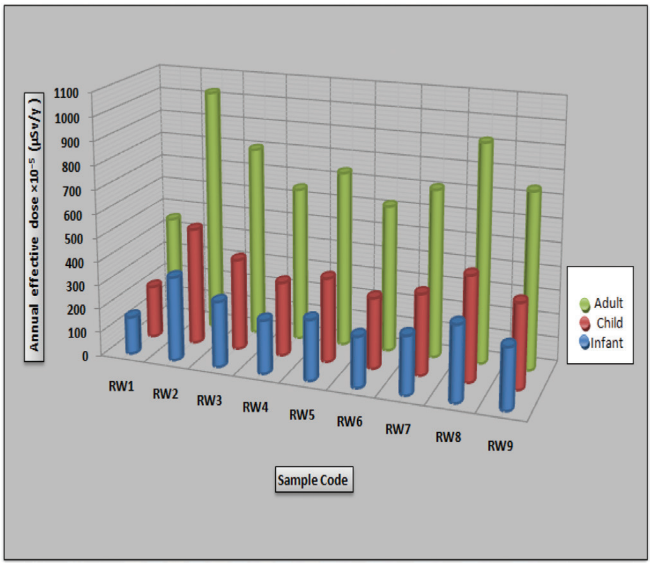

Figure 6: Annual effective dose estimated for members of the public in Tigris river water samples.

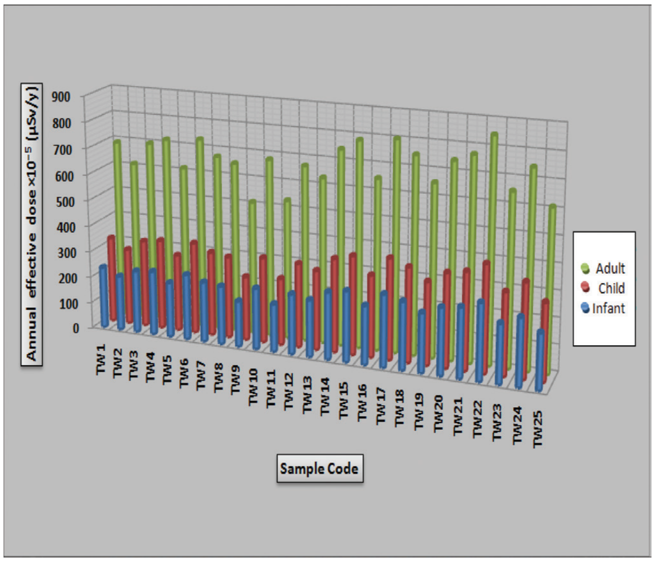

Figure 7: Annual effective dose estimated for members of the public in tap water samples.

Conclusion

In this work, the tritium activity concentrations in drinking water samples collected from the different areas of Al-Amara city were determined using LSC. It can be clearly visible that the evaluated tritium activities in all collected samples of drinking water were below the limit of 100 Bq/L approved by the European Commission and WHO. Moreover, the annual effective dose estimation is done to analyse the radiological risks to all individuals (infants, children, and adults).

Finally, the tritium activity results in state that the drinking water samples consumed in Al-Amara city are radiologically safe for the tritium and do not cause harm to an individual's health. Finally, the results of this study may be useful to set tritium background baselines in these areas and could help to create public awareness about tritium activity concentrations in drinking water and the radiological impact on the dweller's health.

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