

Low-Level Tritium Measurement in Tap Water in Bangkok Area and Annual Dose Estimation

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ARTICLE INFO

Received: 15 Mar 2022
Received in revised: 28 Apr 2022
Accepted: 4 May 2022
Published online: 8 Jun 2022
DOI: 10.32526/ennrj/20/202200066

Keywords:

Liquid scintillation counter/
Tritium/ Tap water/ Minimum
detectable activity/ Annual
effective dose

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ABSTRACT

Monitoring of tritium concentration in tap water is an essential tool to determine the effective dose received from tap water. Liquid scintillation counting (LSC) is a widely used technique for determining tritium in water. Due to the very low activity of tritium in tap water, its detection requires a high-efficiency LSC with the lowest minimum detectable activity (MDA). Low-level tritium analysis in tap water were performed in two LSC models using conventional distillation techniques. The optimal conditions with the lowest MDAs were applied to determine the tritium concentration in the tap water distributed from two main sources located in the Bangkok metropolitan region: Bang Khen and Maha Sawat water treatment plants (WTPs). Twenty-six tap water samples were collected from petrol stations located around both water treatment plants. The results revealed that the amount of tritium in the tap water around the Bangkhen WTP was between 1.88-2.63 Bq/L with an average of 2.28 ± 0.28 Bq/L, whereas those from the Maha Sawat WTP were between 2.01-2.69 Bq/L with an average of 2.44 ± 0.26 Bq/L, which are far below the World Health Organization's (WHO) guideline limit (10,000 Bq/L) for drinking water. The annual effective dose (AED) for infants, children and adults obtained from tap water samples around the Bangkhen WTP were 0.010, 0.014, and 0.030 μ Sv/year, respectively, and those from the Maha Sawat WTP were 0.011, 0.015, and 0.032 μ Sv/year, respectively, which are far below the WHO's guideline limit (100 μ Sv/year).

1. INTRODUCTION

Tritium is a radioactive isotope of hydrogen that emits low-energy beta rays and has a half-life of 12.3 years (Nayak et al., 2019). Tritium can be formed naturally from the interaction of atmospheric nitrogen and oxygen atoms with high-energy cosmic rays (Popoaca et al., 2014), artificially via nuclear reactions (Duliu et al., 2018), and as a by-product of the operation of nuclear reactors (IAEA, 2010). From the mid-1950s to the early 1960s, artificially made tritium was widely dispersed during above-ground testing of nuclear weapons and has been decreasing ever since (USEPA, 2021). Today, tritium emission from nuclear power plants is a major occupational exposure pathway that impacts workers' health. A recent study reported high concentrations of tritium in the urine of

nuclear reactor workers in China, resulting in higher internal radiation exposure caused by tritium compared to the general population (Chen et al., 2021). Tritium, both naturally occurring and artificially made, is usually in gaseous form for under controlled conditions, but when combined with oxygen is either in a liquid state, known as tritiated water (T_2O), or partially formed tritiated water (HTO) with high mobility in the environment (UNSCEAR, 2016; CNSC, 2010). Tritium may pose a health risk if ingested through drinking water, food, inhalation, or absorbed through the skin in large quantities (Matsumoto et al., 2021). The World Health Organization (WHO) has established guidelines for tritium in drinking water at 10,000 Bq/L. This represents the concentration of tritium that, if present

Citation: Sudprasert W, Phattanasub A, Srimork P, Iamlae S, Wongpaiboonsuk P, Wongwechwinit P. Low-level tritium measurement in tap water in Bangkok area and annual dose estimation. Environ. Nat. Resour. J. 2022;20(5):455-464.
(<https://doi.org/10.32526/ennrj/20/202200066>)

in consumed drinking water, would result in an individual dose of 0.1 mSv (WHO, 2017). Therefore, measuring the amount of tritium in drinking water relative to established criteria is an important factor in determining the quality of water used for consumption.

Liquid scintillation counting is a conventional method for the measurement of low-level tritium in environmental samples (L'Annunziata et al., 2020; Theodorsson, 1999). The liquid scintillation counter (LSC) was first introduced in the 1950s and has continued to evolve for over 60 years. Hou (2018) recently reviewed the major developments of LSC's measurement techniques regarding instrumentation, methodology, and its applications in past decades. One major improvement regarding LSC instrumentation and methodology is the commercialization of triple-to-double coincidence ratio (TDCR) in liquid scintillation counting (Cassette and Bouchard, 2003), which has become a routine method for the measurement of beta-emitting nuclides in samples with varying quench levels. The first commercial TDCR-based LSC was introduced in 2008 by Hidex Oy, followed by two more types: the Hidex 300SL Super Low level and Hidex 600SL (Hou, 2018). For analyzing tritium in water samples, the detection limit can be improved by several techniques, such as reducing the quenching effect by using a commercially available tritium column filled with cation and anion exchange resins to purify water samples prior to tritium analysis. Another method with the lowest detection limit is the use of electrolysis to enrich tritium in water samples; however, the main disadvantage is the long analysis time (Hou, 2018). Other possible methods include using a new type of ultra-low background LSC system (e.g., AccuFLEX LSC-LB7, HITACHI ALOKA, Japan) with a 145 mL vial (Feng et al., 2020), or an ultra-low LSC (e.g., Quantulus 1220, PerkinElmer, USA) with commonly used 20 mL counting vials (Varlam et al., 2009).

Since the amount of tritium-in T_2O or HTO form-is present in the tap water samples at very low levels, it is, therefore, necessary to use a highly sensitive LSC with the lowest minimum detectable activity (MDA, Bq/L) for measuring tritium. Numerous recent studies have been especially focused on optimizing the counting performance for tritium analysis (Feng et al., 2020; Arun et al., 2019; Erchinger et al., 2017; Grahek et al., 2016). The results revealed that the main parameters affecting MDA were the volume ratios between the water samples and

the scintillation cocktails, types of cocktails, types of LSC vials, and counting time. The optimal measurement conditions vary across laboratories, depending on available equipment and tools. A recent study determined tritium activity concentrations in tap, well, and spring water samples from Mersin province in Turkey using a Packard TriCarb 2900TR LSC system (Karataşlı et al., 2017). The activity concentration of the tritium measured in water samples varied from <1.9 to 14.1 ± 1.0 Bq/L with an average of 6.2 ± 0.6 Bq/L. From literature review, there was only one study on the measurement of tritium activity concentration in tap water collected from different regions in Thailand using electrolytic enrichment and low background LSC (AccuFLEX LSC-LB7) with a sensitivity of less than 1 Bq/L (Rittirong et al., 2019). The tritium concentrations in tap water were in the range of 0.41-0.75 Bq/L. There are no research data on the concentration of tritium activity in tap water collected in the Bangkok metropolitan area. This research aimed to determine the concentration of tritium in tap water distributed from two main suppliers located in the Bangkok metropolitan region, namely, Bang Khen and Maha Sawat WTPs, to assess the radiological hazards arising from the consumption of tritium in tap water. Both WTPs receive raw water from different sources, the Chao Phraya River and the Mae Klong River, through the eastern and western water-supply canals, respectively. They supply tap water throughout Bangkok and surrounding areas. Therefore, the tap water produced by both WTPs represents the water supply that many people living in Bangkok and nearby areas use for their consumption. If tap water contains high concentrations of tritium, it will affect a large population. The important factors for low-level tritium analysis by two LSC instruments, Hidex 600SL and Quantulus 1220, were optimized using a conventional distillation technique and the optimal conditions were applied for measuring tritium in tap water samples. Determining the tritium levels in tap water is not only important for consumer safety but can also be used as an indicator for tritium leakage into the environment from nuclear activities.

2. METHODOLOGY

2.1 Sampling location

The Bang Khen (13.88165° , 100.55285°) and Maha Sawat (13.80973° , 100.41017°) WTPs are located 14.87 km north-east and 11.37 km north-west of Bangkok City, respectively. The sampling sites

were 26 petrol stations located around the WTPs, which supplied water to those petrol stations. The water supply samples were collected in February 2020. The geographic coordinates and map of the sampling locations are illustrated in **Table 1** and **Figure 1**, respectively.

2.2 Collection of water samples

Before sample collection, the tap was cleaned by washing the outside and inside of the faucet end with detergent and water and running it for at least 1 min. A 1 L Nalgene bottle and cap were rinsed twice by thoroughly shaking with tap water to flush and drain any contaminants that may have remained in the bottle. The tap water samples were then collected in the bottle to approximately 80% of the bottle volume.

2.3 Sample preparation

One hundred milliliters of the water sample were mixed with 0.5 g sodium hydroxide (Merck, Germany) and 0.1 g potassium permanganate (Merck, Germany) in a 500 mL round flask to oxidize the organic matter (ISO, 2019) and then distilled at 80–100°C. The first 20 mL of the distilled water was discarded, and the next 30 mL was collected (USEPA, 2019).

Table 1. Geographic coordinates of each sampling site

Sample code	Latitude (degree)	Longitude (degree)
BK1	13.85829	100.56803
BK2	13.89087	100.56803
BK3	13.93459	100.60911
BK4	13.93285	100.56958
BK5	13.86676	100.59108
BK6	13.85504	100.63265
BK7	13.84508	100.56572
BK8	13.80334	100.57800
BK9	13.83091	100.52550
BK10	13.82551	100.53104
BK11	13.91842	100.51613
BK12	13.89707	100.51545
BK13	13.84234	100.51856
MW1	13.81403	100.41250
MW2	13.82222	100.43685
MW3	13.80233	100.44860
MW4	13.83318	100.41459
MW5	13.87567	100.41240
MW6	13.86511	100.41029
MW7	13.78333	100.41557
MW8	13.78472	100.46134
MW9	13.76198	100.44424
MW10	13.78893	100.34289
MW11	13.77138	100.48790
MW12	13.78341	100.49165
MW13	13.79425	100.50666

BK: Bang Khen; MW: Maha Sawat

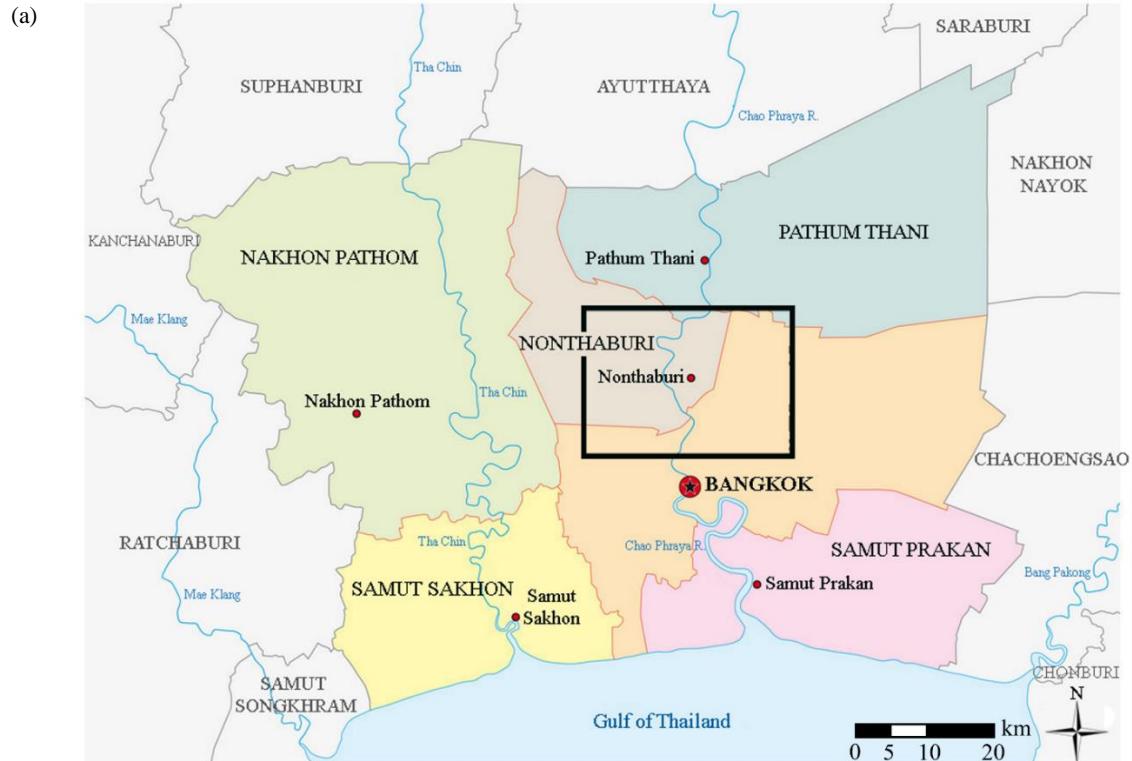


Figure 1. Maps of (a) sampling location in Bangkok metropolitan area and (b) 26 sampling sites generated using Google Maps; yellow pins represent 13 sites around Bang Khen WTP, and red pins represent 13 sites around Maha Sawat WTP.

(b)

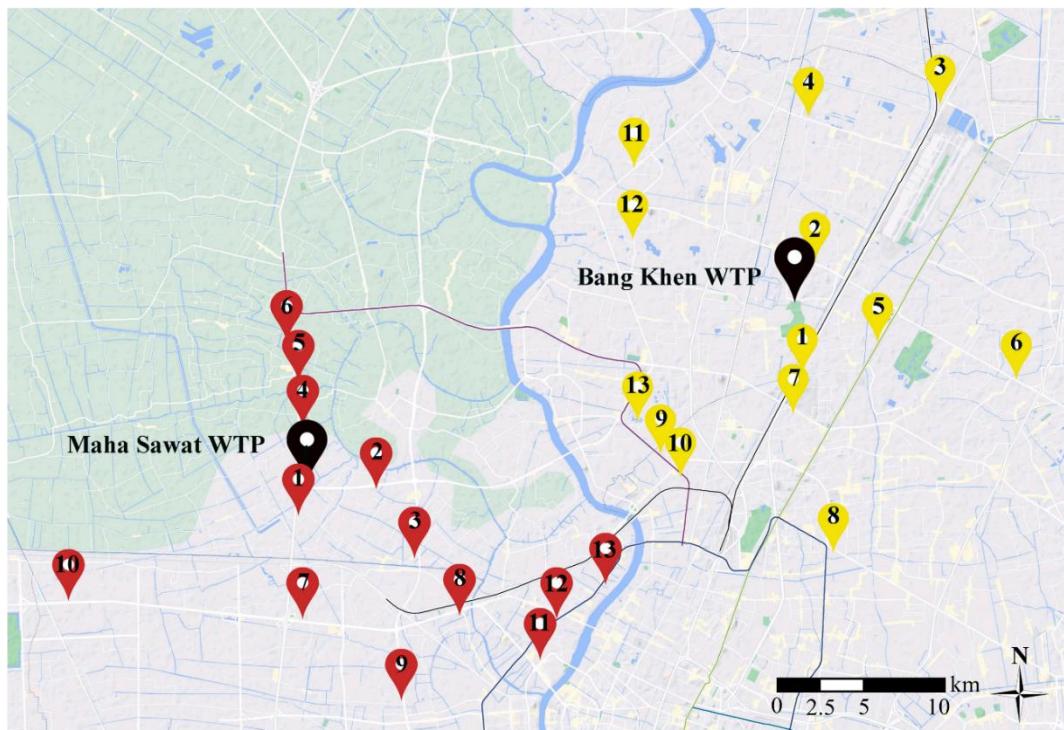


Figure 1. Maps of (a) sampling location in Bangkok metropolitan area and (b) 26 sampling sites generated using Google Maps; yellow pins represent 13 sites around Bang Khen WTP, and red pins represent 13 sites around Maha Sawat WTP (cont.).

2.4 Optimization of the counting performance

2.4.1 Quench curve generation

Two LSCs, the Hidex 600SL automatic (LabLogic Systems, UK) and Quantulus 1220 (PerkinElmer, USA) were used in this study. The efficiency of each tritium measurement was determined by generating a quench correction curve, which is the relationship between the counting efficiency and quench index parameters (QIPs) i.e., TDCR for Hidex and sample channel ratio (SCR) for Quantulus. The quenched tritium standard set (Eckert and Ziegler Analytics, USA) was measured by both LSCs for 60 min, with three measurements per vial. The results were used to generate quench curves, which were subsequently used for the counting efficiency and absolute activity or disintegrations per minute (DPM) calculations (Hou, 2018).

2.4.2 Sample-to-cocktail ratio

The effects of the sample-to-cocktail ratios on the MDA values were determined. The distilled water samples were mixed with the Ultima Gold LLT scintillation cocktail (Perkin Elmer, USA) in 20 mL LSC antistatic high-density polyethylene (HDPE) vials by varying the sample-to-cocktail volume ratio as follows:

- Hidex: 1:14, 2:13, 3:12, 4:11, 5:10, 6:9, and 7:8 mL
- Quantulus: 8:12, 9:11, and 11:9 mL

The vials were shaken well for 2 min and kept in the dark for 24 hours to reduce chemical quenching prior to LSC analysis. Tritium activity was counted for 180 min for Hidex and 480 min for Quantulus.

2.4.3 Vial type and counting time

The effects of vial type and counting time on MDA values were determined by using the optimal sample-to-cocktail ratio 6:9 for Hidex and 11:9 for Quantulus and comparing polyethylene vials and low potassium glass vials with the following counting times:

- Hidex: 60, 120, 180, 360, 540, 720, and 1,440 min
- Quantulus: 60, 120, 240, and 480 min

2.5 Tritium measurements

Measuring tritium in the tap water samples was performed using the optimum conditions obtained from the above experiments. The distilled tap water, tritium-free water and standard tritiated water were added to three LSC vials, followed by the scintillation cocktail. The vials were inverted several times to achieve adequate mixing. The samples were counted

using the Hidex 600SL and Quantulus 1220 LSCs after allowing 24 h of adaptation in the dark. The TDCR outputs obtained from the Hidex were automatically calculated as the absolute sample activity (DPM), whereas the SCR outputs obtained from the Quantulus were manually converted to the counting efficiency. The tritium activity concentration, the annual effective dose and MDA values were then calculated using Equations (1), (2), and (3), respectively.

2.6 Calculations

Using the SCR values, the counting efficiency was determined from the quench curves. The tritium activity concentration, A (Bq/L), was then calculated using Equation (1) (Karataşlı et al., 2017):

$$A \left(\frac{\text{Bq}}{\text{L}} \right) = \frac{R_s - R_b}{E \times V \times 60} \quad (1)$$

Where; R_s and R_b are the count rate of the sample and the background (cpm), respectively, E is the counting efficiency, and V is the sample volume (L).

The annual effective dose, AED ($\mu\text{Sv}/\text{year}$) due to the ingestion of tritium in the drinking water samples was calculated using Equation (2) (Pintilie-Nicolov et al., 2021):

$$AED = A \times CF \times CR \times 10^6 \quad (2)$$

Where; A is the tritium activity concentration (Bq/L), CF is the dose coefficient ($1.8 \times 10^{-11} \text{ Sv/Bq}$ for tritium), and CR is the consumption rate of drinking water (250, 350, and 730 L/year for infants, children, and adults, respectively) (WHO, 2017).

The MDA (Bq/L) was calculated based on the Currie equation (Currie, 1968) using Equation (3):

$$MDA \left(\frac{\text{Bq}}{\text{L}} \right) = \frac{\left(\frac{2.71}{T_s} \right) + 3.29 \left(\frac{R_b}{T_b} + \frac{R_b}{T_s} \right)^{1/2}}{E \times V \times 60} \quad (3)$$

Where; R_b is the count rate of the background (cpm), T_s is the counting time of the sample (min), T_b is the counting time of the background (min), E is the counting efficiency derived from the quench curve, and V is the sample volume (L).

3. RESULTS AND DISCUSSION

3.1 Quench curve generation

Since the counting efficiency is affected by the level of quenching in the sample, in order to determine the absolute sample activity, it is necessary to establish

a quench correction curve to measure the quenching level in the samples. Theoretically, quench correction can be performed by one of the following methods: (1) internal standard method, (2) sample spectrum method, (3) external standard method, and (4) direct DPM method (L'Annunziata et al., 2020). These techniques allow for determining the detection efficiency of a particular sample and for converting the count rate (CPM) to the disintegration rate (DPM). In our study, the quench curves were obtained from a set of 10 quenched tritium standards containing constant tritium activity (222,540 Bq) with varying levels of quenching using the TDCR method for Hidex 600SL (Gudelis et al., 2017) and the SCR method (L'Annunziata et al., 2020) in the selected counting windows of 1-175 and 176-350 channels for Quantulus 1220. Figure 2(a) shows the quench curve for tritium, which shows a correlation between the TDCR value and the counting efficiency, which is a second-order polynomial curve given by the equation $y = -0.5856x^2 + 1.5468x - 0.098$ ($R^2 = 0.9993$), where x is the TDCR value and y is the counting efficiency. Thereafter, the counting efficiency of an unknown sample can be determined from the measured TDCR value of the sample (Hou, 2018). The DPM value is then automatically calculated after the quench curve is generated and stored on a computer. Likewise, Figure 2(b) shows the quench curve obtained from Quantulus, showing a correlation between the SCR value and the counting efficiency, which is a logarithmic curve given by the equation $y = -0.114\ln(x) + 0.4427$ ($R^2 = 0.9816$), where x is the SCR value and y is the counting efficiency. The tritium activity concentration is calculated from Equation (1) using the counting efficiency derived from the above equation.

3.2 Sample-to-cocktail ratio

The water sample to scintillation cocktail ratio is particularly important for measuring tritium using LSC (Lin et al., 2020). Therefore, the optimal sample-to-cocktail ratio needs to be determined. From the measurement results, it was found that the ratio between the volume of water and the Ultima gold LLT liquid scintillation cocktail influenced the MDA value, in which it was found that the MDA decreased when the sample-to-cocktail ratio increased (Figure 3). For Hidex, the lowest MDA was found at 6.9 Bq/L with a 7:8 sample-to-cocktail ratio, when samples were counted for 180 min (Figure 3(a)). However, the counting efficiency was found to be the highest at a 6:9 ratio. Therefore, a 6:9 sample-to-cocktail ratio was

determined as the optimal ratio and was used for further experiments. For Quantulus, the lowest MDA was found at 1.02 Bq/L with an 11:9 sample-to-

cocktail ratio when samples were counted for 480 minutes (Figure 3(b)).

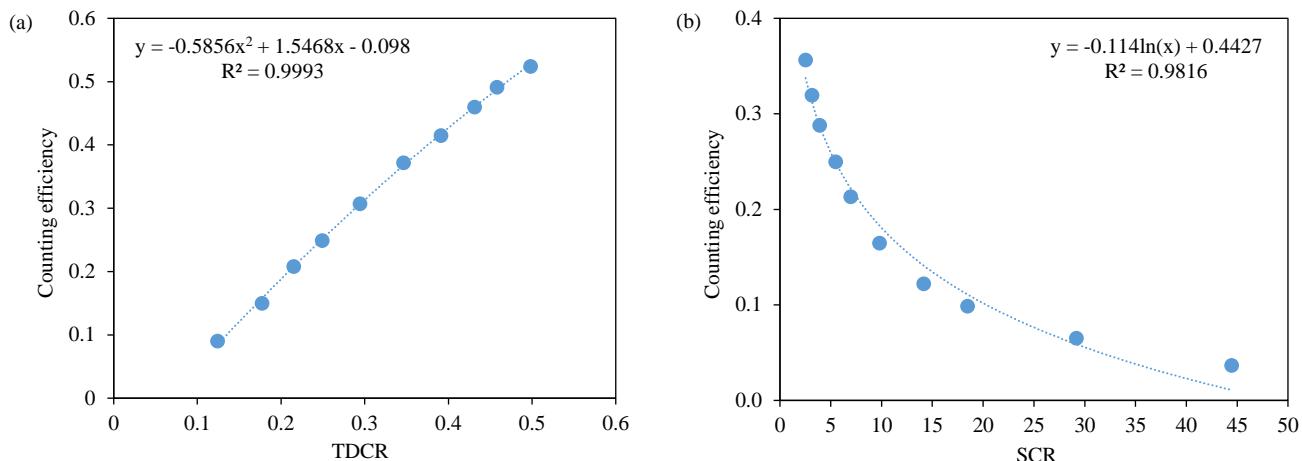


Figure 2. Quench curves for tritium counting using (a) TDCR and (b) SCR values as the quench index parameter

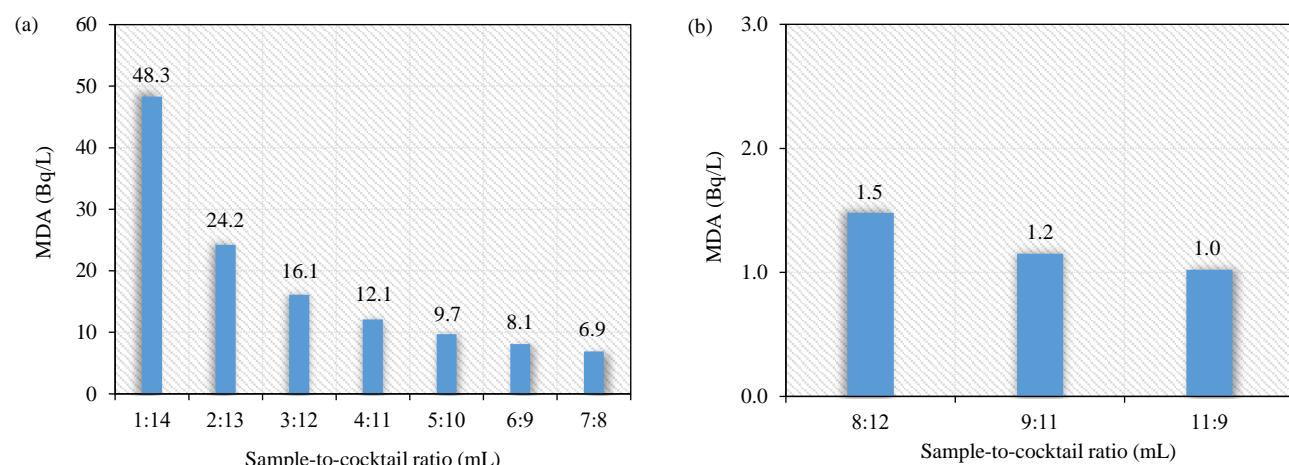


Figure 3. Change in MDA values in each sample-to-cocktail ratio: (a) Hidex and (b) Quantulus

3.3 Vial type and counting time

The effects of LSC vial type and counting time on MDA values were investigated. The results showed that the type of LSC vial and counting time influenced the MDA values, as shown in Figure 4. Plastic vials were found to have lower MDA values than glass vials for both LSCs. These results are consistent with a recent study (Arun et al., 2019) since plastic vials have lower background values than glass vials. The higher background value in the glass vial could be from K-40. Although LSC vials are often made of borosilicate glass, they are lower in potassium; however, their performance in the ${}^3\text{H}$ energy region was significantly affected by radioactive residues in the glass (Perkin Elmer, 2021). Therefore, plastic vials are appropriately

used for measuring low-level tritium activity in environmental samples.

The counting time had an inverse relationship with the MDA value. The lowest MDA was found at the maximum counting time used in this study, i.e., 1,440 min for Hidex and 480 min for Quantulus. Therefore, counting times of 1,440 and 480 min were used for low-level tritium measurements in tap water samples using Hidex and Quantulus LSCs, respectively.

3.4 Optimum conditions

The optimum conditions for measuring tritium in tap water samples are summarized in Table 2. The lowest MDA of tritium measurements in tap water

samples was found to be 2.7 and 1.0 Bq/L using Hidex and Quantulus LSCs, respectively. This result is consistent with previous studies by [Hanslik et al. \(2005\)](#), [Palomo et al. \(2007\)](#), [Varlam et al. \(2009\)](#), and [Jakonić et al. \(2014\)](#). The detection limits of standard or ultra-low background LSCs are around 1-3 Bq/L for non-enriched water samples. The Hidex 600SL LSC achieved a higher MDA of 2.7 Bq/L with the following optimum conditions: a sample-to-cocktail ratio of 6:9 counted for 1,440 min in a plastic vial.

Although Hidex cannot determine the tritium content in tap water because it is below the MDA value, more importantly, the new MDA value, obtained by our laboratory, is significantly lower than the original 9.6 Bq/L using a 5:10 sample-to-cocktail ratio in a plastic vial. As tritium is a radioactive substance with limited health effects, due to its low energy emissions, measuring low levels of tritium in the environment is more useful for monitoring the impact of nuclear activities than public health concerns.

Table 2. The lowest MDA values obtained from the optimal conditions of the two LSCs

LSC	Sample-to-cocktail ratio	Vial Type	Counting time (min)	MDA (Bq/L)
Hidex 600SL	6:9	Plastic	1,440	2.7
Quanlutus 1220	11:9	Plastic	480	1.0

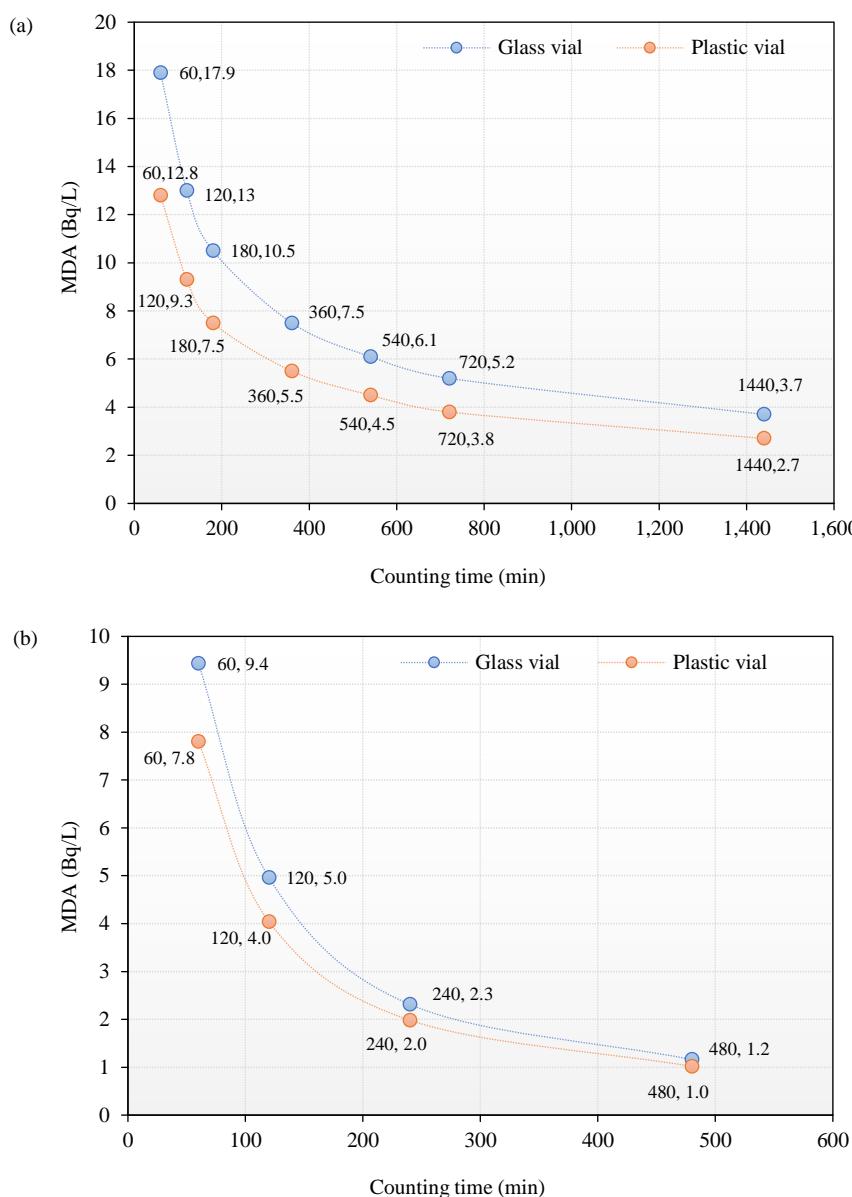


Figure 4. MDA values with different vial types and counting times: (a) Hidex and (b) Quantulus

3.5 Tritium concentration in tap water

Measuring tritium using the optimum parameters of both LSCs showed that Hidex was unable to detect tritium due to the tritium content being lower than its MDA value of 2.7 Bq/L, while the Quantulus, with a lower MDA of 1.0 Bq/L, was able to measure tritium. The results of the tritium concentrations with their respective MDAs from each sampling location are shown in Figure 5. The tritium concentration in tap water around the Bang Khen WTP location was between 1.88-2.63 Bq/L with an average of 2.28 ± 0.28 Bq/L, whereas those from the Maha Sawat WTP were between 2.01-2.69 Bq/L with an average of 2.44 ± 0.26 Bq/L. The average amount of tritium in tap water around the two WTPs was less than 10,000 Bq/L, which is a WHO guideline for drinking water quality (WHO, 2017). Our results are comparable with a recent study by Pédehontaa-Hiaa et al. (2020), which reported that the average tritium concentration in tap water in Lund, Sweden, measured in 2018-2019, was 1.5 ± 0.6 Bq/L (MDA=1.2 Bq/L). It is noteworthy that the average tritium content in tap water supplied from the Bang Khen WTP was not

significantly different from that supplied from the Maha Sawat WTP. Although the raw water source used in the production of the water supply of the two plants differs, the amount of tritium was not impacted in any way. The Bang Khen WTP uses raw water from the Chao Phraya River through the eastern water supply canal to the WTP, which is a total distance of about 18 km to produce tap water. The Maha Sawat WTP uses raw water from the Mae Klong River above the Mae Klong dam through the western water supply canal to the WTP, which is a total distance of 107 km (Metropolitan Waterworks Authority, 2020).

Based on our preliminary data, if tritium concentrations were assessed in water samples across the country, they could then be used as a basis for comparing tritium concentrations and assessing the future impact on nuclear power plants. However, if the tritium content is below 1.0 Bq/L, the electrolytic enrichment method is required since it provides an extremely low MDA value (Walova et al., 2020; Lin et al., 2020), but is more laborious than the conventional distillation method (Hou, 2018).

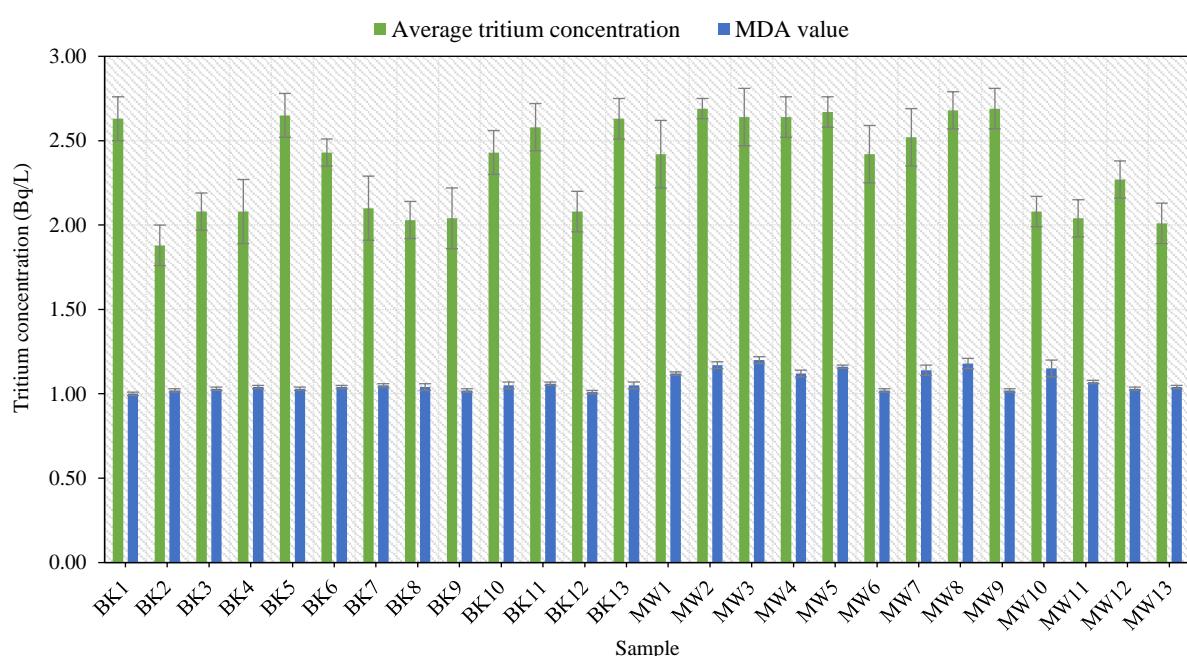


Figure 5. Average tritium concentrations in tap water collected from 26 stations around Bang Khen (BK) and Maha Sawat (MW) water treatment plants compared to their MDA values

3.6 The assessment of the annual effective dose

To assess the effective dose resulting from the ingestion of tritium in tap water, the AED per capita were calculated in infants, children and adults. The average AEDs from each sampling site are shown in

Figure 6. The AED for infants, children and adults obtained from tap water samples around the Bangkhen WTP were 0.010, 0.014, and 0.030 $\mu\text{Sv}/\text{year}$, respectively, and those from the Maha Sawat WTP were 0.011, 0.015, and 0.032 $\mu\text{Sv}/\text{year}$, respectively,

which are far below the WHO's guideline limit (100 $\mu\text{Sv}/\text{year}$). Our results are comparable to a previous study by [Karataşlı et al. \(2017\)](#), which reported that the AED values due to the intake of the tritium in

natural water samples for infants, children, and adults in Turkey were averaged 0.024, 0.034, and 0.070 $\mu\text{Sv}/\text{year}$, respectively.

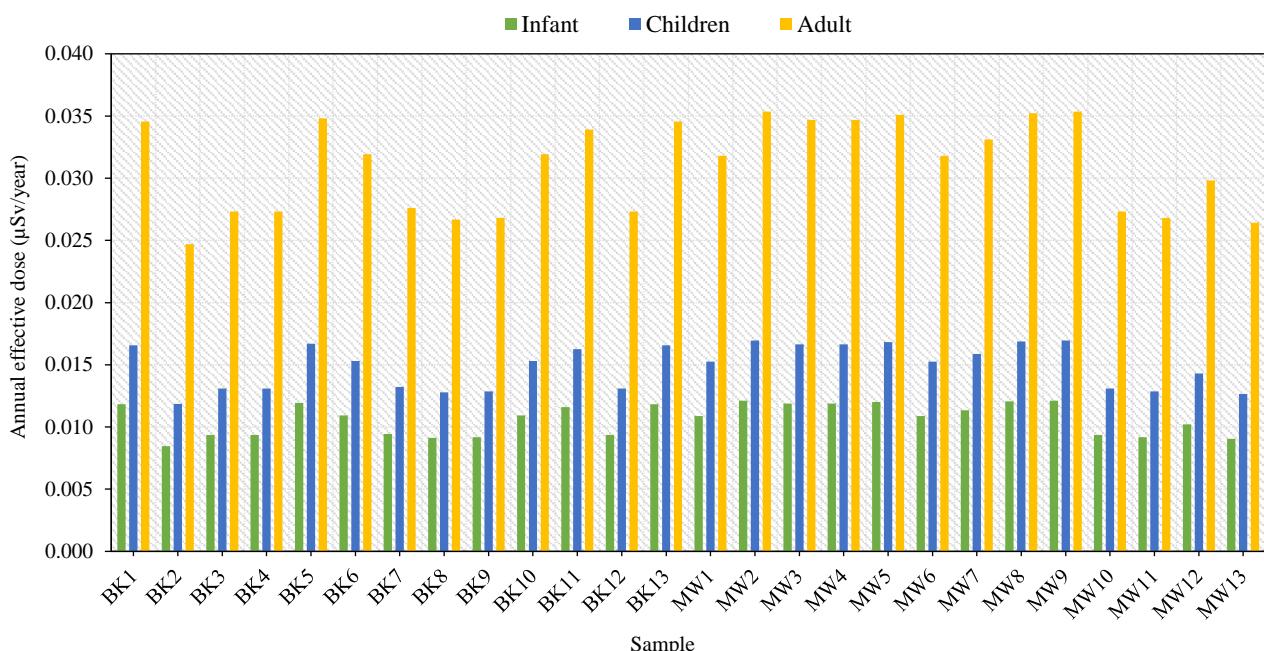


Figure 6. Annual effective dose in infants, children and adults derived from the average tritium concentration in tap water collected from 26 stations around Bang Khen (BK) and Maha Sawat (MW) water treatment plants

4. CONCLUSION

Due to the low level of tritium in tap water samples, the optimum conditions for Quantulus 1220 were successfully applied for directly measuring tritium without pre-enrichment which are simple and economical. The concentration of tritium in tap water around the Bangkhen WTP was between 1.88-2.63 Bq/L with an average of 2.28 ± 0.28 Bq/L, whereas those from the Maha Sawat WTP were between 2.01-2.69 Bq/L with an average of 2.44 ± 0.26 Bq/L, which is far below the WHO's guideline limit (10,000 Bq/L) in drinking water. The annual effective dose resulting from the ingestion of tritium in tap water from both WTPs are far below the WHO's guideline limit (100 $\mu\text{Sv}/\text{year}$). As tritium is a radioactive substance with limited health effects due to its low energy emissions, measuring low levels of tritium in the environment is more useful in monitoring the impact of nuclear activities than public health concerns. This improvement could support environmental monitoring programs throughout the country.

ACKNOWLEDGEMENTS

This work was supported by the Thailand Institute of Nuclear Technology (Public Organization)

through the TINT2U program and the Faculty of Science, Kasetsart University. The authors are grateful to the Radioactive Waste Management Center, Thailand Institute of Nuclear Technology (Public Organization), for their technical and facility support.

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