Determination of tritium levels in tap waters collected from various regions in Thailand using liquid scintillation counting

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Abstract. Tritium can replace a hydrogen atom in water molecules since oxygen atom can react with tritium atoms to form water molecules the same way as for hydrogen atoms, potentially posing hazardous to consumers. This study aims to measure tritium activity concentrations in tap water samples collected from different regions in Thailand, (Phayao, Srisaket, Chanthaburi, Trang and Bangkok) in November 2018. The measurements of tritium activity concentrations were carried out by using electrolytic enrichment and low background liquid scintillation counting (AccuFLEX LSC-LB7), which had a sensitivity of less than 1 Bq/L. The data results of regional distribution of a tendency were discussed according to latitudes and longitudes of the collecting location. The measured tritium concentrations in tap water were in the range of 0.41 – 0.75 Bq/L. Specifically, the results showed that the tritium activity concentrations depended on location, with the highest value obtained from a tap water sample collected from the northern part of Thailand (Phayao). The results could be used as the baseline of tritium levels in tap water in each region of Thailand before building or operating the nuclear facilities such as the first nuclear power plant, fuel reprocessing or heavy water production facilities in the future.

1. Introduction

Tritium ($^3$H) is one of the existing natural radioisotopes of hydrogen. Most natural tritium occurs when secondary cosmic rays (fast neutrons) interact with nitrogen atoms in earth’s atmosphere ($^1$H, $^3$H, $^12$C). An average production rate of tritium is estimated to be 0.5 – 1.9 atoms/cm² s [1]. Tritium is transported through the hydrological cycle from earth’s atmosphere to surface since tritium can interact with oxygen atoms to form water molecules, the same way as hydrogen atoms interact with oxygen atoms. Water molecules that have tritium atoms are called tritiated water (HTO). Other sources of tritium are anthropogenic activities such as nuclear reactors and atmospheric thermonuclear tests. These anthropogenic sources play an important role in tritium activity concentrations in environment, especially, the atmospheric thermonuclear tests that took place in 1953 – 1963, in which both of United States of America and Soviet Union tested their nuclear weapons in the northern hemisphere at Marshall Island and Kazakhstan, respectively. The treaty signing to stop the atmospheric thermonuclear tests was
done in 1963. The maximum concentration of tritium in precipitation sample collected from various regions of Australia in that year was 160 tritium unit (TU) [2] (1 TU corresponds to 1 tritium atom in $10^{18}$ hydrogen atoms and equal to 0.118 Bq/L [3]). Moreover, average annual measured tritium in precipitation samples from Ottawa (Canada) and Vienna (Austria) in the same year was around 2900 TU and 3177 TU, respectively due to several thermonuclear tests in areas. In Thailand, tritium measurement had been started since 1968, in which the concentration of tritium in rain water samples collected in Bangkok was about 46.01 TU [4]. Additionally, not only the atmospheric thermonuclear tests are the significant sources of tritium in environment but also other nuclear activities such as nuclear reactors or nuclear reactor accidents could release the tritium to environment. For example, on 11 March 2011, the latest nuclear reactor accident at Fukushima Dai-ichi nuclear power plants (Japan), maximum tritium concentration was found in the first rainfall at Tsukuba, which was almost 30 times higher than pre-accident levels [5]. However, tritium is a radioisotope continuously decays to $^3$He by emitting low energy beta particle with half-life of 12.3 years [6]. Thus, tritium activity concentrations in environment decrease with time. Nowadays, Tadros et al. [2] has been recording the tritium activity concentrations measured in Australia monthly, since 1960 in cooperation with the International Atomic Energy Agency (IAEA). The results show that present levels of tritium concentrations in precipitation samples are approximately 2 – 3 TU. For northern hemisphere, according to IAEA GNIP database, the average tritium in precipitation samples in 2016 was about 13.67 TU.

Environmental tritium can be transported through hydrological cycle (from atmosphere $\rightarrow$ rain $\rightarrow$ river or lake $\rightarrow$ consumed tap water). Many countries in the world consume or use tap water for cooking and drinking especially in Thailand. However, the limit of tritium activity concentration in drinking water is 10,000 Bq/L according to Guidelines for Drinking-water Quality suggested by WHO [7], which is much higher than the tritium levels found in tap water samples.

The aim of this study was to determine the tritium activity concentration in tap water collected from various regions of Thailand and to study the effects of latitudes and longitudes on the tritium levels in tap water. The result was expected to be used as the baseline of tritium levels in tap water in Thailand before operating the first nuclear facilities in the near future.

2. Methodology

2.1. Tap water sample collection

Five tap water samples were collected in November 2018 using glass bottles with screw cap from various regions in Thailand including Phayao (Mueang Phayao), Srisaket (Uthumpornphisai), Bangkok (Chatuchak), Chanthaburi (Laem Sing) and Trang (Mueang Trang). These provinces are in the northern, northeastern, middle, eastern and southern part of Thailand, respectively (Figure 1.).

2.2. Electrolytic enrichment for tritium

The present tritium concentration in environment is extremely low even in the precipitation samples, comparing with the values obtained in 1960s, which had several atmospheric thermonuclear tests. To determine the tritium levels in environmental samples such as precipitations, ground water or tap water, the electrolytically enrichment process is required. The electrolytic apparatus used in this research was solid polymer electrolyte type with that separated produced hydrogen and oxygen gases, and with electronic cooler to prevent evaporation of water sample made by National Institute for Fusion Science (NIFS), Japan.
Two hundred milliliters of tap water samples were enriched to get a 10 mL final volume. The enrichment operated with 10.0 A current could enrich approximately 3 g of water samples per hour. Thus, it took almost 3 days to enrich from 200 mL to 10 mL of water samples according to Faraday’s factor. The enrichment factor was determined using the spike method (high concentration tritiated water) and was calculated by the equation (1) [8].

\[ R = \frac{T_f \cdot V_f}{T_i \cdot V_i} \]  

Where R represents the tritium enrichment factor of electrolytic apparatus, \( T_i \) and \( T_f \) represent tritium concentration in measured samples before and after enrichment, respectively. \( V_i \) and \( V_f \) represent volume of sample before and after enrichment, respectively. Tritium enrichment factor can be determined by the measurement of high concentration tritiated water with and without enrichment. The tritium concentration in tap water sample before enrichment could be calculated through equation (2) which was rearrangement of equation (1).

\[ T_i = \frac{T_f}{\left( \frac{V_i}{V_f} \right) \cdot R} \]  

2.3. Liquid Scintillation Counter

The final volume of enriched water samples was mixed homogenously with 10 g of Ultima GOLD™: PerkinElmer, in 20 mL polyethylene vials (Zinsser Analytic). The tritium counting was done by using a

Figure 1. Tap water sampling site in Thailand. Including Phayao, Srisaket, Bangkok, Chanthaburi and Trang.

Figure 2. Electrolytic enrichment for tritium with oxygen and hydrogen separation.
low background liquid scintillation counter (Hitachi AccuFLEX LSC LB-7) with triple coincidence photomultiplier tube and an active-passive shield, which provides sensitivity for tritium measurement of less than 1 Bq/L. To avoid a peculiar scintillation caused by chemiluminescence and photonolouscence, the cocktails were kept in dark at a low temperature condition (10°C) for 24 hours and flowed the ultra-pure nitrogen gas to purge the radon gas in the measurement chamber. The measurement time was preset for 100 minutes with 10 repeats, resulting in the total measurement time of 1,000 minutes. Tritium contents in water samples and their uncertainties could be calculated using equation (3). The H-gross of measurement sample (Ni) divided by measurement time (t) were used to calculate the tritium activity concentrations by subtracting tritium-free water samples, collected at Tohno underground mine that was older than 1 million years. The Ti (Bq/L) of each measuring sample was calculated following its condition using the measurement efficiency (E) and volume sample (V) in the unit of % and mL, respectively.

\[
T_i \left( \frac{\text{Bq}}{L} \right) = \left[ \frac{N_i}{t_i} - \frac{N_b}{t_b} \right] \pm \left( \frac{N_i}{t_i} + \frac{N_b}{t_b} \right)^{1/2} \cdot \frac{1}{E} \cdot \frac{1}{V} \cdot \frac{1}{60} \cdot 10^5 \tag{3}
\]

For quenching correction, the external standard channel ratio (ESCR) was used. \(^{133}\)Ba as a gamma ray source was used to calculate the efficiency of each measurement via correction curve (Figure 3.). In order to obtain the correction curve, the ESCR calibration samples were prepared by adding standard tritiated water with the activity of 9.95 \times 10^2 Bq/g on March 22\textsuperscript{nd} 2016, Ultima GOLD\textsuperscript{TM} (PerkinElmer) and distilled water as a quenching agent following the condition in table 1.

### Table 1. Calibration values used to obtain the quenching correction curve.

| HTO (g) | H\(_2\)O (g) | Scintillation Cocktail (g) | Total (g) | Disintegration per second [DPS] | Count per second [CPS] | Efficiency (EFF) [%] | External Standard Channel Ratio (ESCR) |
|---------|-------------|---------------------------|----------|-----------------------------|------------------------|----------------------|-------------------------------------|
| 0.997   | 5.940       | 12.710                    | 19.647   | 734.145                     | 95.440                 | 13.00                | 11.61                               |
| 0.996   | 6.920       | 11.760                    | 19.676   | 733.375                     | 89.838                 | 12.25                | 10.97                               |
| 1.013   | 7.950       | 10.750                    | 19.713   | 658.514                     | 79.549                 | 12.08                | 10.69                               |
| 0.999   | 10.940      | 8.800                     | 20.739   | 736.357                     | 74.446                 | 10.11                | 9.01                                |
| 0.999   | 9.920       | 9.000                     | 19.919   | 732.080                     | 69.767                 | 9.53                 | 8.16                                |

### 3. Results and Discussion

The tritium concentrations in tap water samples collected from various regions of Thailand were enriched using electrolytic enrichment for tritium. The tritium enrichment factor was calculated by measuring the concentration of tritiated water with and without the enrichment and calculated the volume reduction factor of enriched sample using initial sample volumes from 100 mL and 200 mL. Our electrolytic enrichment apparatus had 0.4137 enrichment factor, which could enrich 3.1 times and 8.2 times from initial sample volumes at 100 mL and 200 mL, respectively (Table 2.).

The volume reduction factor of each sample was calculated by the ratio of initial and final volumes (V/\(V_i\)). The tritium concentration before enrichment was calculated through equation (2) using volume reduction factor, measured tritium concentration in enriched sample (T\(_i\)) and enrichment factor (R). The results of tritium concentration in tap water were in the range of 0.41 – 0.75 Bq/L (Table 3.). The highest concentration was from Phayao, which is located in the northern part of Thailand.
Figure 3. Quenching correction curve for 20 mL polyethylene vial.

Table 2. Enrichment factor of electrolytic enrichment apparatus.

| Sample                  | $T_f$ (Bq/L) | Volume reduction factor ($V_i/V_f$) | Enrichment Factor ($R$) | $T_f/T_i$ |
|-------------------------|--------------|------------------------------------|-------------------------|-----------|
| HTO without enrichment  | 113.6515 (T_i) | -                                  | -                       | -         |
| HTO 200 mL with enrichment | 932.2937   | 21.4385                            | 0.3826                  | 8.2031    |
| HTO 100 mL with enrichment | 353.3274   | 6.9906                             | 0.4447                  | 3.1089    |

Table 3. Tritium concentration in tap water before enrichment.

| Sample            | $T_f$ (Bq/L) | Volume reduction factor ($V_i/V_f$) | $T_i$ (Bq/L)    |
|-------------------|--------------|------------------------------------|-----------------|
| HTO without enrichment | 113.65     | -                                  | 113.65 ± 2.54  |
| HTO 200 mL with enrichment | 932.29     | 21.4385                            | 105.12 ± 2.85  |
| HTO 100 mL with enrichment | 353.33     | 6.9906                             | 122.17 ± 2.58  |
| Bangkok           | 4.76         | 21.8555                            | 0.53 ± 0.02    |
| Phayao            | 6.71         | 21.7085                            | 0.75 ± 0.02    |
| Trang             | 4.85         | 19.8748                            | 0.41 ± 0.02    |
| Chanthaburi       | 4.47         | 19.9860                            | 0.54 ± 0.02    |
| Srisaket          | 5.44         | 23.3891                            | 0.57 ± 0.02    |

The results of tritium concentrations in tap water were discussed by latitudes and longitudes of the sampling sites (Figure 4.). The measured tritium concentrations in tap water collected in November 2018 were found to be the highest in the north (Phayao) and the lowest in the south (Trang). When comparing the results from this work with the previous work reported from samples collected from eastern part of China in July to September 2014 and August 2015 which in the range of 0.02 – 1.42 Bq/L [9], they were similar (within the same order of magnitude). Tritium concentrations versus latitudes and longitudes of the collecting locations also showed the trend with the highest value in the north and northwest and the lowest in the south and southeast. Additionally, Tadros et al. [2] studied the effects of latitude trends by
long-term recording of tritium in precipitation samples collected from various region of Australia in 1960 to 2010. The result showed that the tritium concentrations in precipitation samples increased with latitudes, which was same with the result from tritium study in the atmosphere by Gonfiantini et al. in 1991 [10] This behavior was due to the incoming cosmic ray in the polar region. These support the higher tritium concentration in the north of China and Thailand for upper hemisphere. On the other hand, the results from previous measurement in eastern part of China also showed that tritium concentration in tap water were higher in the northwest.

![Tritium Concentration vs. Latitude](image1.png)

**Figure 4.** Regional distribution of tritium concentration in 5 tap water samples collected from various region of Thailand

4. Conclusion
Tritium activity concentrations in tap water were in the range of 0.41 – 0.75 Bq/L. The highest concentration was found in the northern part of Thailand (Phayao), which was related to the highest latitude among all samples. This could be explained that tritium concentration in atmosphere was increased by latitude, causing by incoming cosmic-ray. This preliminary study provided tritium reference data in tap water samples and could be used for future environmental studies in Thailand.

5. References
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