

Tritium Concentration in Natural Spring Water Collected at Hirosaki, Japan^{*)}

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In order to understand the regional property of tritium concentration in natural water at Hirosaki City, Aomori Prefecture Japan, natural spring water samples were collected from 15 sites in 2016 and tritium concentration was measured by low-level tritium counting procedure using a commercially available solid polymer electrolyte (SPE) tritium enrichment system. The limit of detection of this procedure was approximately 0.06 Bq/L. Findings showed tritium concentrations in natural spring water samples ranged from 0.30 to 0.55 Bq/L with mean value of 0.44 ± 0.08 Bq/L. These results were within the range of reported concentrations in rain and surface and shallow groundwater in Japan including other places of Aomori Prefecture. The presently obtained data could be considered as the background level of the Hirosaki area. The annual effective dose would be negligibly small compared with the annual effective dose limit of 1 mSv, if the committed effective dose equivalent from drinking water to residents was calculated using the highest tritium concentration data.

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1. Introduction

Tritium (^3H ; $T_{1/2} = 12.3$ y) is the radioisotope of hydrogen and it decays to ^3He ; the main sources of environmental tritium are natural, although there are some artificial sources [1]. Nuclear fusion releases a large amount of energy when light atomic nuclei fuse together by thermonuclear reactions [2]. In the future, electrical power generation using deuterium-tritium (D-T) nuclear fusion reactors would require a large inventory of tritium as fuel (kg) [3]. Therefore, nuclear fusion facilities will also be important sources of artificial tritium. The deuterium plasma experiment using the Large Helical Device (LHD) has been conducted at the National Institute for Fusion Science (NIFS) since March 2017 to investigate high-temperature plasma physics and hydrogen isotope effects in research leading towards the realization of fusion energy. It has made great achievements in plasma research [4]. After the deuterium plasma experiment started, the LHD has been producing small amounts of neutrons and tritium atoms by fusion reactions.

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To understand the impact from nuclear fusion facilities on the environment and for public acceptance of artificially sourced tritium in the environment, it is important to survey the background tritium concentration in environmental water samples [5–8]. Data for some areas have already been reported [9, 10]. However, concentration levels in recent years have been low and require an enrichment procedure [1].

Here, calibration of a tritium enrichment system is performed. Then, tritium concentration in natural spring water at Hirosaki, Japan is determined and reported to clarify the regional background level.

2. Materials and Method

The spring water samples were collected at 15 locations in Hirosaki City, Aomori Prefecture, located in the northern part of Honshu Island in the Japanese archipelago. (Fig. 1). Details of sampling locations were reported by Yamada *et al.* [11]. The basic geological information of this area was also reported [12]. The bedrock of the Hirosaki area is mainly composed of the Jurassic accretionary complex and Cretaceous plutonic rocks, and it is covered

in parts by the Neogene system and sediments of post-Pleistocene and volcanic products. The aquifers in this area are located in the Neogene Pliocene to Quaternary Pleistocene.

Briefly, all samples were drinkable water, and samples were collected using 1 L polyethylene bottles in 2016. At that time, pH, electrical conductivity (EC) and water temperature were measured using a pH meter (AS-711, HORIBA, Japan), EC meter (B-771, HORIBA, Japan) and thermometer (CT-220, CUSTOM Corporation, Japan), respectively. After bringing the water samples back to the laboratory, they were kept in a refrigerator for preparation. First, approximately 900 mL of sample water was distilled to remove impurities. Next, 800 mL of the distilled water sample was electrolyzed to about 65 mL to enrich its tritium content using the commercially available solid polymer electrolyte (SPE) tritium enrichment system, TRIPURE®(XZ001, De Nora Permelec Ltd.) [13]. An illustration of this enrichment system is shown in Fig. 2. In this experiment, the electrolysis was done at a constant current of 50 A for 40 h and then at 20 A until the final volume of sample reached approximately 65 mL.

The 65 mL of tritium-enriched sample was distilled, and 50 mL of this distilled water was mixed with 50 mL of liquid scintillation cocktail (UltimaGold LLT, PerkinElmer) in a 145 mL low diffusion polyethylene vial with an inner Teflon coating [14]. Each polyethylene vial

with the enriched sample and liquid cocktail was shaken to make a homogeneous solution that was stored overnight inside a low background liquid scintillation counter (LSC: LSC-LB5, Nihon RayTech, Japan) after over 5 days. The next day tritium radioactivity measurements were done for 1,000 min using the LSC, and counting efficiencies were determined using a secondary standard tritium solution (118.17±0.39 Bq/L, reference date: December 20, 2020) which was certified against a reference tritium solution (SRM 4361C, NIST). The limit of detection (LOD) of the LSC-LB5 was approximately 0.3 Bq/L.

3. Results and Discussion

3.1 Evaluation of enrichment factor

The tritium enrichment factor (Z_T) of the TRIPURE system is defined using the following equation [15]:

$$Z_T = \frac{T_f}{T_i} = \left(\frac{V_i}{V_f} \right)^{\left(1 - \frac{1}{\beta_a}\right)}, \tag{1}$$

where V_i and V_f are initial and final water volumes, T_i and T_f are initial and final tritium concentrations, and β_a is apparent separation factor, respectively. The tritium enrichment factor (Z_T) was obtained ten times using diluted secondary standard tritium solution. The extreme values were rejected from the data obtained by the statistical test ($k-\sigma$). As a result, the final volume of sample was approximately 68 mL and 4.93±0.39 was the estimated average Z_T with standard deviation. The LOD of this procedure with tritium enrichment is approximately 0.06 Bq/L. The relative expanded uncertainty of this procedure ($k = 2$) was less than 11%. The tritium analysis conditions for this procedure are summarized in Table 1.

3.2 Tritium concentration in natural spring water

Tritium concentrations obtained in natural spring water samples collected at Hirosaki, Japan are shown in Fig. 3. Tritium concentrations for the samples and other parameters are listed in Table 2 with ^{222}Rn concentrations reported by Yamada *et al.* [11]. The air temperatures of sampling days ranged from 14.7 to 28.0°C [11]. On the other hand, water temperature ranged from 7.4 to 21.7°C. Sampling points 3 and 4 had similar temperatures (difference about 5°C). This result indicated that the aquifer of these points was shallow. Tritium concentration in the

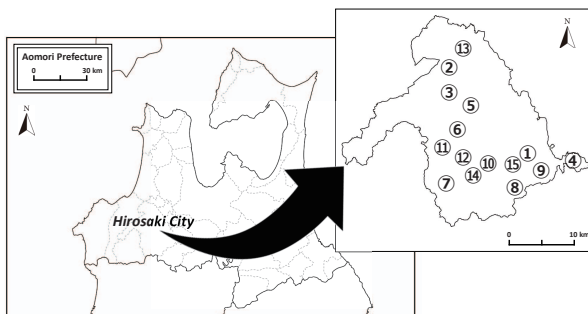


Fig. 1 Location of sampling points.

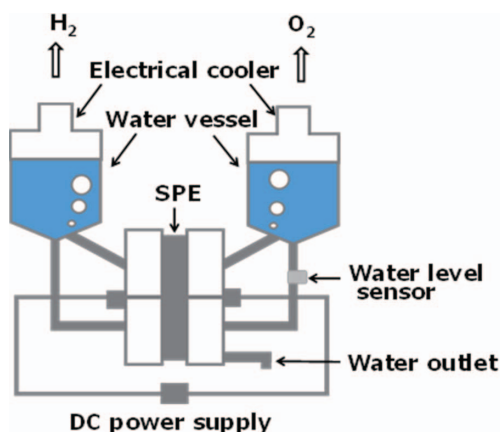


Fig. 2 Overview of SPE electrolytic system.

Table 1 The analytical conditions of tritium for this procedure.

Final volume (mL)	approximately 68 mL
Enrichment factor (EF)	4.93±0.40
Limit of Detection (LOD)	0.06 Bq/L
Relative expanded uncertainty ($k = 2$)	< 11 %

samples ranged from 0.30 to 0.55 Bq/L with mean value of 0.44 ± 0.08 Bq/L. The highest values were observed at points 1 and 7, and the lowest one was observed at point 10. No regional characteristics were observed in the small catchments in this study.

There are some recent reports about tritium concentrations in natural water in Japan including places near the present study area [16–18]. For example, tritium concentration in rain samples from Hirosaki during 2018 to 2020 was reported to range from 0.28 to 1.20 Bq/L [19]. Hasegawa *et al.* [20] reported that tritium concentrations in surface and shallow groundwater from Rokkasho Village located on the Pacific Ocean side of Aomori Prefecture in 2009 - 2012 ranged from 0.03 to 0.57 Bq/L. Outside of Aomori Prefecture, the tritium concentrations in rain samples from Sapporo City located on the large northern island of the Japanese archipelago during 2015 to 2019 and Toki City located in the central part of the main island during 2014 to 2020 ranged from 0.24 to 1.59 and 0.1 to 0.86 Bq/L, respectively [21, 22]. On the other hand, tritium concentration in rain samples from Chiba City located

near the metropolitan Tokyo area have been reported since 1973, and the recent Chiba City concentration range during 2019 - 2021 was 0.09 to 0.63 Bq/L [23]. A summary of tritium concentration ranges is shown in Fig. 4. The present results were within the range of concentration in precipitation and surface and shallow groundwater in Rokkasho. Tritium in rain is known to have temporal and spatial trends influenced by season, latitude, continental-maritime origin of air masses, and altitude. And natural spring water and surface and shallow groundwater are affected by rainwater. As a result, the present data were almost the background level in northern Japan which includes Hirosaki City.

Tritium is one of the low-energy beta emitter radionuclides and should only be considered for internal exposure. The annual effective dose for tritium was determined according to the following equation [11]:

$$D = D_{WI} \times D_{CF} \times Y \times C_A, \tag{2}$$

where D is the annual effective dose (mSv) from the consumption of tritium in the water, C_A is the tritium concentration in the water (Bq/L), D_{WI} is the daily consumption of water (the value was estimated to be 2 L/d as provided by WHO guidelines), D_{CF} is the dose conversion factor of ^3H for adults, (1.8×10^{-11} Sv/Bq provided by the ICRP) and Y is the ingestion period (365 d). The maximum annual

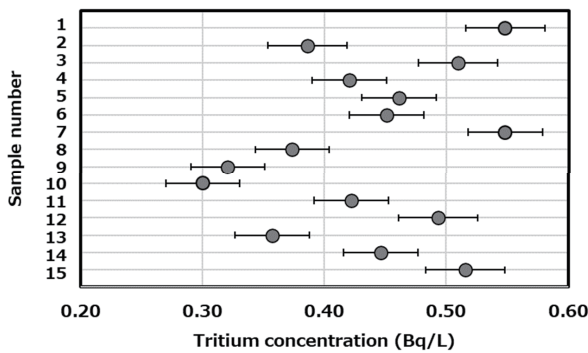


Fig. 3 Tritium concentration in natural spring water at Hirosaki, Japan.

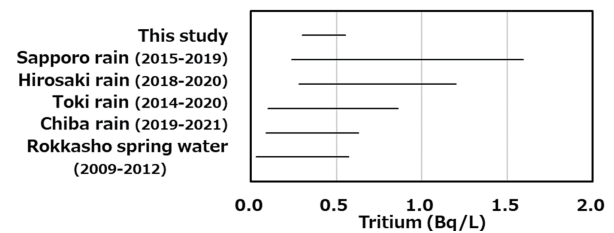


Fig. 4 Range of tritium concentrations in water samples collected in Japan.

Table 2 The tritium and ^{222}Rn concentrations in sample water and other parameters.

No.	Sampling Date	pH	EC ($\mu\text{S}/\text{cm}$)	Water temp. ($^{\circ}\text{C}$)	Tritium Conc. (Bq/L)*	^{222}Rn Conc. (Bq/L)**
1	4 Aug. 2016	6.5	250	15.2	0.55 ± 0.06	16.5 ± 1.0
2	17 Aug. 2016	7.2	153	14.7	0.39 ± 0.05	6.7 ± 0.2
3	18 Aug. 2016	6.4	86	19.7	0.51 ± 0.06	13.9 ± 0.9
4	21 Aug. 2016	6.2	111	21.7	0.42 ± 0.05	11.5 ± 0.7
5	22 Aug. 2016	6.7	89	7.4	0.46 ± 0.05	5.3 ± 0.5
6	25 Aug. 2016	6.5	100	6.9	0.45 ± 0.05	6.3 ± 1.0
7	29 Aug. 2016	5.7	220	12.5	0.55 ± 0.06	16.6 ± 0.6
8	5 Sep. 2016	6.6	119	9.9	0.37 ± 0.05	11.0 ± 0.8
9	7 Sep. 2016	6.6	68	9.7	0.32 ± 0.04	26.7 ± 1.1
10	8 Sep. 2016	6.1	185	11.3	0.30 ± 0.04	8.0 ± 0.7
11	15 Sep. 2016	5.8	140	10.3	0.42 ± 0.05	18.8 ± 0.8
12	16 Sep. 2016	6.2	198	9.7	0.49 ± 0.06	18.6 ± 1.2
13	19 Sep. 2016	5.9	164	9.9	0.36 ± 0.05	7.9 ± 0.3
14	26 Sep. 2016	6.2	240	10.2	0.45 ± 0.05	7.7 ± 1.0
15	11 Sep. 2016	5.9	320	16.2	0.52 ± 0.06	13.8 ± 0.3

*Measured value \pm counting error **From [11]

effective dose due to the ingestion of tritium was calculated as $0.007 \mu\text{Sv}$. This value was negligibly small compared with 1 mSv , which is the index of the annual dose limit for the public. Yamada *et al.* [11] reported that ^{222}Rn and ^{226}Ra concentrations in the same water samples ranged from < 4 to $14 \mu\text{Sv}$; these are also important radionuclides for internal dose assessment from drinking water. When the maximum values of both calculated values were combined to make a conservative estimate, the dose was negligibly small. Tritium concentration is one of the useful tools to understand the local water environment. In the future, it is planned to use these data to understand the regional property and to estimate the residence time of spring water.

4. Conclusion

To understand the regional background concentration of tritium in natural spring water in northern Japan, samples were collected at 15 locations in Hirosaki City Aomori Prefecture. Tritium concentration in the water samples ranged from 0.30 to 0.55 Bq/L with mean value of $0.44 \pm 0.08 \text{ Bq/L}$. There was no local geographical property in this small catchment. The present results were within the range of reported concentrations in rain and surface and shallow groundwater including other places of Aomori Prefecture. Tritium in rain is known to have temporal and spatial trends influenced by season, latitude, continental-maritime origin of air masses, and altitude. And natural spring water and surface and shallow groundwater are affected by precipitation. As a result, it seems that the present data are the background level of the Hirosaki area. When the committed effective dose equivalent from drinking water to residents was calculated using the highest data, it was negligible small compared with annual effective dose limit of 1 mSv .

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The authors declare that they have no conflict of interest.

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