

Tritium Concentration of Inland Water at Rokkasho Village, Aomori Prefecture

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This study adds new data points for 2024 to the existing time series of tritium concentrations in inland water in Rokkasho Village, Aomori and evaluate its relevance for environmental safety. Rokkasho has hosted major nuclear facilities, including nuclear power plants and a nuclear fuel reprocessing facility, and been considered a candidate site for a fusion facility in Japan. Since tritium is both a fuel for fusion and a waste product from nuclear reprocessing, background concentration data is essential for environmental monitoring during future operations. The most recent measurements in this area were conducted by Hasegawa *et al.* in 2006 during final testing of the nuclear fuel reprocessing facility with actual fuel and now are outdated. In this study, water samples were collected from 10 sites in October 2024. Tritium concentrations were measured using low-level tritium counting method with a commercially available solid polymer membrane enrichment system. Tritium concentration ranged from 0.21 to 0.37 Bq L⁻¹ and the mean value is 0.29 ± 0.06 Bq L⁻¹, which is lower than previously reported in Rokkasho. Based on this mean concentration, the estimated internal dose was 3.84 × 10⁻⁶ mSv year⁻¹ which is negligible compared with the public dose limit (1 mSv year⁻¹). These results indicate that the background tritium levels remain very low, and continuous monitoring is important to provide reliable baseline data for future environmental assessments.

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1. Introduction

Tritium is a radioactive hydrogen isotope with a half-life of 12.3 years, emitting low-energy beta radiation with a maximum energy of 18.6 keV [1]. Tritium is produced naturally in the atmosphere through interactions between cosmic rays and nitrogen and oxygen [2]. In addition, there are anthropogenic sources, including nuclear power plant operation, spent or used fuel reprocessing, nuclear weapon testing during the 1950s until the early 1960s [3], and releases from fusion facilities. It can exist in various chemical forms in the environment. The most common form is tritiated water (HTO), followed by gaseous tritium (HT) and bound in organic matter in the biosphere called organically bound tritium (OBT) [4]. Although it emits low-energy beta radiation and has been considered a minor radiological hazard, it can disperse rapidly in the atmosphere, making it important to monitor continuously, particu-

larly in areas surrounding nuclear facilities. Once released, these forms undergo atmospheric dispersion and deposition onto soil, water, and vegetation. HTO is of particular importance for environmental and radiological assessments because it is incorporated into the hydrological cycle and can be absorbed by the human body through inhalation, skin absorption, and ingestion of drinking water or food [5]. Internal dose evaluation is especially related to HTO because the human body consists largely of water. In addition, the dose coefficient of HTO is approximately 10,000 times greater than that of HT [6, 7]. For these reasons, the present study focuses on tritiated water (HTO) in inland waters.

The Rokkasho area, where a fuel reprocessing plant is located, therefore represents a significant site for monitoring tritium levels and understanding its environmental behavior. The reprocessing plant is planned to start full operation around 2026, and preparations for its start are currently ongoing. During the test operation in 2006, tritium was released from the nuclear reprocessing plant into the atmosphere and the ocean.

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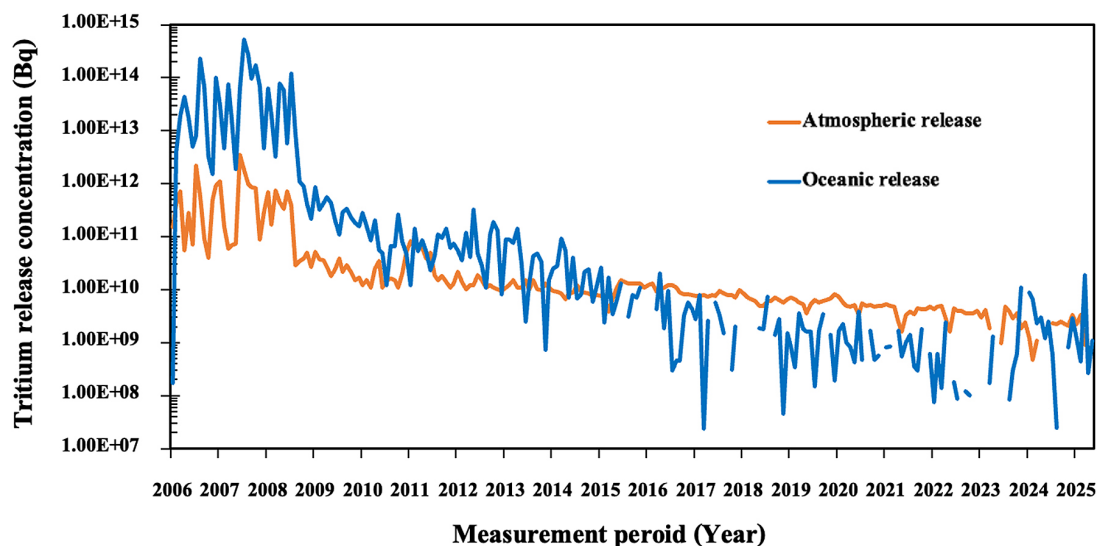


Fig. 1. Monthly Release to Atmosphere and Ocean during 2006 to 2025.

As shown in Fig. 1, during the early operation (2006–2008), atmospheric tritium releases ranged from 10^{11} – 10^{12} Bq month⁻¹, whereas releases to the ocean were higher, ranging from 10^{13} – 10^{14} Bq month⁻¹. After 2009, atmospheric releases generally decreased and stabilized at approximately 10^{10} Bq month⁻¹ (range from 10^9 – 10^{11} Bq month⁻¹). Conversely, oceanic releases displayed larger temporal variability and show an overall decreasing trend from 2009 to 2025, with a range of approximately 10^7 – 10^{12} Bq month⁻¹.

Although the amount of tritium that is released from the reprocessing plant has been measured, limited information on recent environmental tritium concentrations in inland water has been reported since the last testing phase, which began in March 2006 and used real spent nuclear fuels [8, 9]. To address this gap, we collected additional samples of inland water in October 2024.

In addition, this area has been considered a prospective site for the JA DEMO (Japan's fusion DEMO) demonstration reactor [10]. While a fusion facility is not currently in operation, it is expected that large amounts of tritium will need to be managed there [11]. Establishing baseline data on environmental tritium levels is important for evaluating future environmental monitoring.

This study aims to provide current background information on tritium levels in the Rokkasho area, compare them with previous results reported by Tonosaki *et al.* [12], Ueda *et al.* [13], and Hasegawa *et al.* [14] and evaluate the internal dose from ingestion to assess whether the levels remain within safe limits [15]. The present results serve as baseline data for future environmental assessments and will be useful for evaluating potential changes in tritium levels associated with the fusion-related facilities in the region. These findings may support environmental monitoring efforts relevant to governmental agencies and the public.

2. Materials and Methods

2.1 Sampling

Water samples were collected from 10 locations in Rokkasho Village, located on the Pacific Ocean side of Aomori Prefecture, Japan, where a reprocessing plant by Japan Nuclear Fuel Limited was operated (JNFL) [16]. This survey was conducted in October 2024, before the winter season in this area. The sampling locations were selected to be consistent with those in a previous study by Hasegawa *et al.* [14]. At each site, approximately 1,000 mL of water was collected in polyethylene bottles. These sites included both river water and marsh area water, as shown in Fig. 2. Water temperature was measured simultaneously with sample collection.

2.2 Tritium measurement

Before tritium analysis, water quality parameters, including pH and electroconductivity (EC), were measured using a pH meter (AS-711, HORIBA, Japan) and an EC meter (B-771, HORIBA, Japan). Tritium concentrations were determined by using distillation and electrolytic enrichment using a solid polymer electrolyte (SPE) system. The analytical procedure followed this improved SPE system developed by Akata *et al.* [17], which enhances the efficiency of tritium enrichment.

This procedure began with a distillation step to separate the water from chemical contaminants such as salt and organic compounds, processing 600 mL of water per sample. Enrichment was performed using the Tripure system (XZ001, De Nora Permelec LTD, Japan), a SPE specifically designed for tritium enrichment before measurement with a liquid scintillation counter. The diagram of the system is shown in Fig. 3 consisting of two reservoirs with Peltier-type electrical coolers, an electrolysis cell, and a DC power supply [18]. During electrolysis in SPE, the water sample is decomposed to molecular hydrogen and oxygen. At the cathode, hydrogen ions are transported through the SPE membrane, which consists of perfluoro-sulfonic acid are reduced hydrogen gas. This mem-

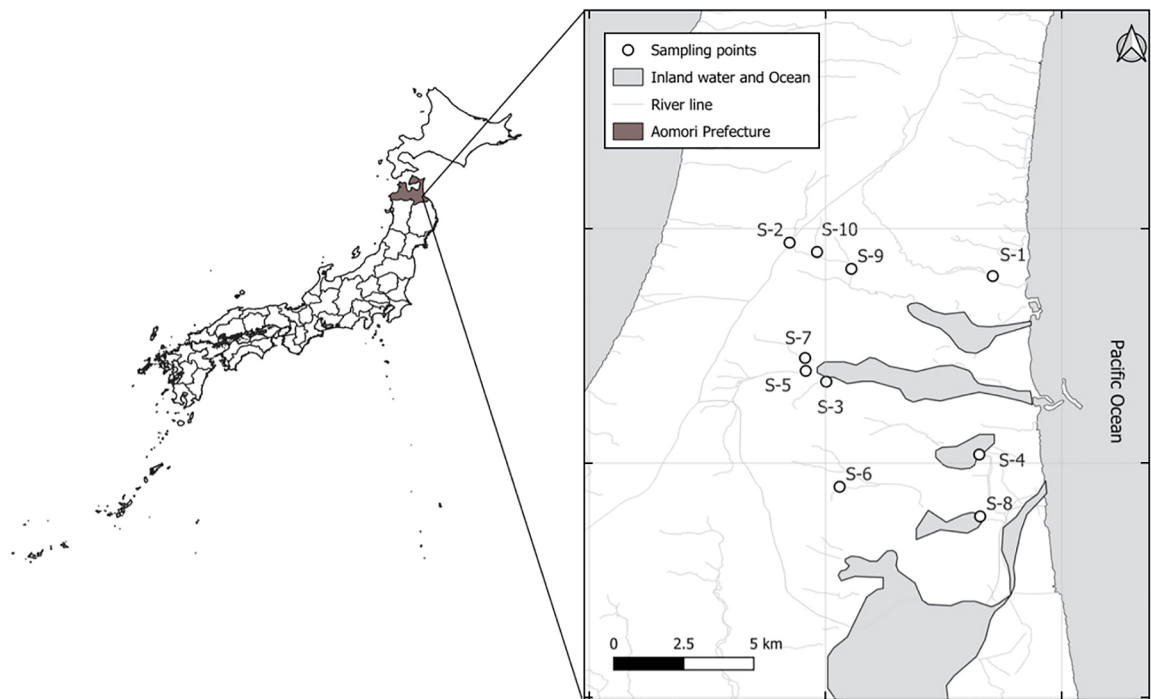


Fig. 2. Location of the sampling points in Rokkasho Village.

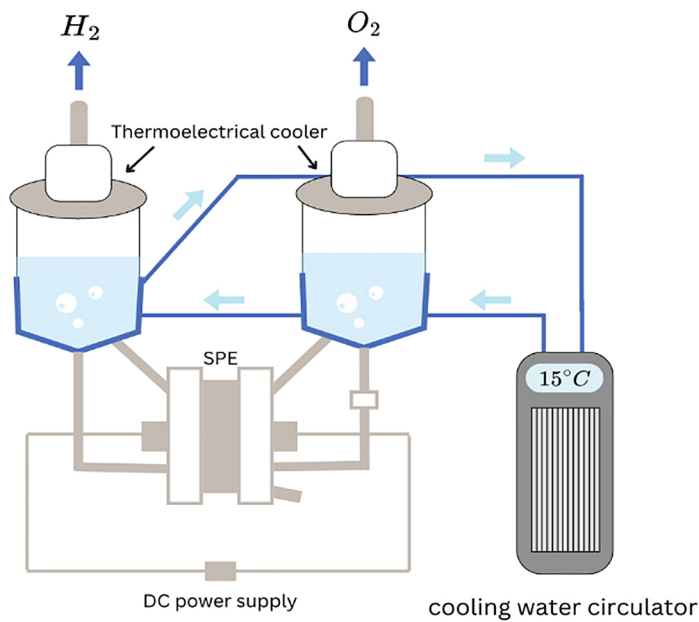


Fig. 3. The schematic diagram of the solid polymer electrolytic (SPE).

brane prevents the mixing of H_2 and O_2 produced by the electrolysis of water [19]. The electrolysis cell is composed of SPE film made of fluorocarbon resin, a porous titanium anode coated with iridium oxide, and a porous SUS316 cathode [18]. This configuration improves stability and safety by ensuring the separation of hydrogen and oxygen gases, thereby reducing explosion risk. To minimize water loss, the gases were released from each side of the water vessel through the thermoelectric cooler. The reservoirs were modified with a double-glass layer to control the sample water temperature. Water used

for cooling, kept at a temperature of $15^{\circ}C$ during the entire process, helped to reduce overheating of the system and stabilize electrolysis effectiveness, which is important since temperature variation can influence isotope separation efficiency due to the isotope effect [20]. These conditions were followed by a second distillation step aimed at further purification of samples.

Electrolysis was performed with an initial water volume of 500 mL post-distillation. The electrolysis was conducted at a constant direct current (DC) of 50 A for 22 hours, followed

by 20 A until the remaining volume was reduced to approximately 55 mL. The relation between tritium concentration and water volume during electrolysis was defined by the following equation:

$$\frac{V_f}{V_i} = \left(\frac{T_f \times V_f}{T_i \times V_i} \right)^\beta. \quad (1)$$

Where T_i and T_f are initial and final tritium concentrations, V_i and V_f are initial and final water volumes, and β is the separation factor of tritium. The separation factor is influenced by isotope fractionation caused by evaporation, internal pressure within the electrical cell, and the vapor pressure of sample water. Therefore, an apparent separation factor β_a , is typically determined experimentally. The tritium enrichment factor (Z_T) was defined by the following equation:

$$Z_T = \frac{T_f}{T_i} = \left(\frac{V_i}{V_f} \right)^{(1-1/\beta_a)}. \quad (2)$$

After enrichment, the final sample was mixed with 50 mL of liquid scintillation cocktail (Ultima Gold LLT, Revvity, US) and transferred in a 145 mL low-diffusion polyethylene vial with an internal Teflon coating to prevent contamination of the sample. The tritium concentration was measured on a low-background liquid scintillation counter (LSC-LB5, ALOKA, Japan). Each sample was measured for a total of 1,000 minutes to assure sufficient accuracy and sensitivity. The relative uncertainty of tritium after enrichment was less than 10% ($k = 2$). Using the SPE system at an initial volume of 500 mL and LSC counting time of 1,000 minutes, the minimum detection level was estimated to be 0.06 Bq L⁻¹. In addition, the estimated Z_T value was 4.93 ± 0.39 , showing an increase in the enrichment factor when compared to conventional methods [21].

2.3 Estimation of internal dose

The annual effective dose (Sv) from ingestion of tritium in water was estimated using the following equation:

$$D = C_A \times D_{WI} \times D_{CF} \times Y. \quad (3)$$

Where D is annual effective dose (Sv) from the ingestion of tritium in the water, C_A is the tritium concentration in the water (Bq L⁻¹), D_{WI} is the daily consumption of water (the value was estimated to be 2 L per day) [21], D_{CF} is the dose conversion factor of tritium for adults (1.8×10^{-11} Sv Bq⁻¹) and Y is the ingestion period (365 days) [22].

3. Results and Discussions

Table 1 presents the tritium concentration measured in inland water samples from Rokkasho Village. Figure 4 compared this study (October 2024) with Hasegawa *et al.* (October 2006) [14] at the same sampling locations in Rokkasho Village. As shown in the figure, tritium concentrations in the current study are systematically lower than those reported by Hasegawa *et al.* (2006) at the same sampling points in Rokkasho Village. The concentrations in 2024 ranged from 0.21 to 0.37 Bq L⁻¹, with an overall mean of 0.29 ± 0.06 Bq

Table 1. Tritium concentration and other parameters of each sampling point.

Sampling point	pH	EC ($\mu\text{S cm}^{-1}$)	Water temperature ($^{\circ}\text{C}$)	^3H (Bq L ⁻¹)
S-1	7.57	144	17.8	0.35 ± 0.02
S-2	7.42	119	17.9	0.27 ± 0.02
S-3	7.40	172	15.6	0.28 ± 0.02
S-4	7.59	147	24.0	0.35 ± 0.02
S-5	7.54	197	16.3	0.27 ± 0.02
S-6	6.94	270	17.0	0.25 ± 0.02
S-7	7.29	270	17.1	0.21 ± 0.02
S-8	9.06	560	24.6	0.37 ± 0.02
S-9	7.49	141	17.3	0.29 ± 0.02
S-10	7.48	149	18.5	0.28 ± 0.02

L⁻¹, whereas the 2006 concentrations ranged from 0.37 to 0.69 Bq L⁻¹.

Among the sampling points, the Tamogi area (S-8, marsh water) showed the highest tritium concentration (0.37 ± 0.02 Bq L⁻¹), while the Muronokubo area (S-7, river water) had the lowest concentration (0.21 ± 0.02 Bq L⁻¹). Focusing specifically on the Futamata River area (S-2, S-9, and S-10) where the Nuclear Fuel Cycle Facilities are located, the tritium concentration ranged from 0.27 to 0.29 Bq L⁻¹. Compared to data from 2006 at nearby locations, which showed concentration ranging from 0.37 to 0.49 Bq L⁻¹, these results indicate a moderate decrease in tritium level. Similarly, Ueda *et al.* [14] found an average tritium content of 0.79 Bq L⁻¹ in the Futamata River during 2001–2004, which is more than twice the level observed in the present study. Tonosaki *et al.* (2000) reported tritium concentrations in the river and lake of Rokkasho during 1993–1994, with values reaching up to 2.0 Bq L⁻¹ [13].

Furthermore, monitoring in Hirosaki City, which is located in the same prefecture, reported tritium concentrations in spring water ranging from 0.28 to 1.20 Bq L⁻¹ [17]. The concentrations in this study fall within this reported background range, indicating that the measured values are consistent with typical environmental levels in northern Japan. In addition, recent environmental monitoring data from other regions in Japan, including Niigata Prefecture and Nagasaki Prefecture, reported tritium concentrations within a similar range, which were approximately 0.25–0.50 Bq L⁻¹ [23].

To further characterize the observed decline shown in Fig. 5, the environmental half-life of tritium ($T_{1/2,eff}$) was estimated to evaluate the long-term decline of tritium concentrations in inland water [21]. The estimation assumed a first-order exponential decay of tritium concentration using the following equation:

$$\ln C_t = -\lambda_{eff}t + \ln C_0. \quad (4)$$

Where C_0 and C_t are the initial and final tritium concentrations, respectively, (Bq L⁻¹), and λ_{eff} is the effective rate constant of decline in radioactivity concentration, and t is time (year). With this, the effective half-life was obtained from:

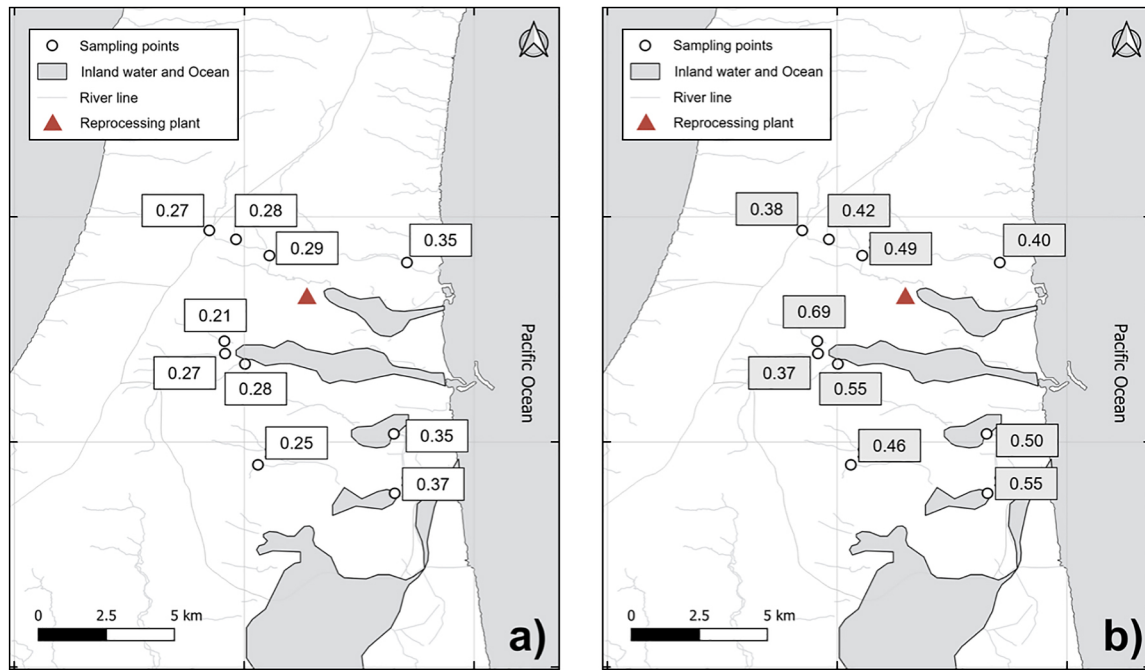


Fig. 4. Distribution of ^3H concentrations (Bq L^{-1}) in inland water samples from Rokkasho Village measured in (a) October 2024 (this study) and (b) October 2006 (Hasegawa *et al.*).

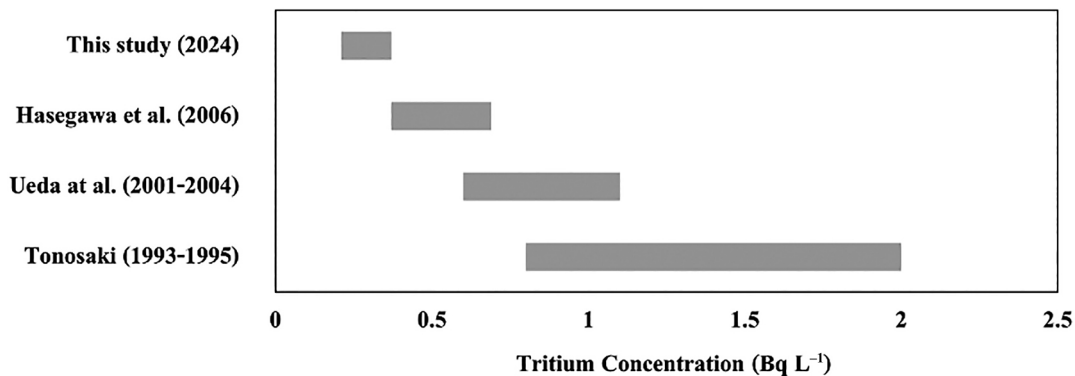


Fig. 5. Range of tritium concentrations in water samples collected in Rokkasho Village [12–14].

$$T_{\text{eff}} = \frac{\ln 2}{\lambda_{\text{eff}}} \quad (5)$$

Using the mean values reported in previous studies and the present results, the calculated of T_{eff} was approximately 9.5 years. This value is slightly shorter than the physical half-life of tritium (12.3 years). This reduction likely reflects the end of active testing with spent nuclear fuel, radioactive decay, and dilution processes in the hydrological system.

As shown in Fig. 1, atmospheric tritium concentration releases during the early test operation period (2006–2008) were 10^{11} – 10^{12} Bq month^{-1} , whereas after 2009 the releases decreased and stabilized at approximately 10^{10} Bq month^{-1} . This presented a reduction of approximately one to two orders of magnitude compared to the peak testing period. The estimated effective half-life found in this study ($T_{\text{eff}} \approx 9.5$ years) showed that the long-term decline of environmental tritium concentration attributable to reprocessing plant operation was

observed. This indicates that environmental processes are actively removing tritium from the local inland system. The measured concentrations remain within the typical environmental background range. Although the releases continued during subsequent years, these releases occur through residual handling activities rather than through active fuel testing. Despite the continued input of low-level releases, the environmental tritium concentrations in inland water showed a declining trend. This suggests that under the present release conditions, the contribution of reprocessing activities to inland water tritium concentration is not significant compared to natural background and hydrological dilution processes. The analytical performance of the measurement method was less than 10% ($k = 2$), corresponding to approximately 0.02 – 0.03 Bq L^{-1} . This study observed variation in tritium concentration (0.21 – 0.37 Bq L^{-1}), corresponding to approximately 0.16 Bq L^{-1} , which clearly exceeds this analytical uncertainty, indicating that the spatial variability reflects environmental differences

rather than measurement variability.

The estimated effective dose from consuming this water, based on the mean tritium concentration, was approximately 3.84×10^{-6} mSv year⁻¹. This value is far below the WHO guideline of 0.1 mSv year⁻¹ for drinking water [24], confirming that current tritium levels in Rokkasho inland waters pose no significant health risk. Nevertheless, continuous monitoring is warranted, particularly due to the presence of the Nuclear Fuel Cycle Facilities and the potential future construction of Japan's demonstration fusion reactor (JA DEMO).

4. Conclusion

The background concentration of tritium in inland waters within the Rokkasho area was generally low. The internal dose from water ingestion was approximately 3.84×10^{-6} mSv year⁻¹, which is lower than 0.1 mSv year⁻¹ drinking water guideline by WHO. Furthermore, the finding also indicated a reduction in tritium concentration compared to previous studies, consistent with its natural radioactive decay and potential environmental dilution over time. Moreover, the results suggested that the current level of tritium release from the reprocessing plant does not produce a measurable increase in inland water tritium concentrations under the present environmental conditions. In particular, these present results provide baseline information that will be useful for environmental assessment of tritium levels when the reprocessing plant or a fusion prototype reactor begins operation.

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