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journal or publication title	Plasma and Fusion Research
volume	11
page range	1305032
year	2016-03-17
URL	http://hdl.handle.net/10655/00012677

doi: 10.1585/pfr.11.1305032



Long-Term Monitoring of Tritium Concentration in Environmental Water Samples Collected at Tono Area, Japan

Naofumi AKATA^{1,2)}, Masahiro TANAKA^{1,2)}, Hiromi KATO¹⁾, Hirokuni YAMANISHI³⁾, Hideki KAKIUCHI⁴⁾, Hiroshi HAYASHI¹⁾, Hitoshi MIYAKE¹⁾ and Kiyohiko NISHIMURA¹⁾

¹⁾National Institute for Fusion Science, Toki, Gifu 509-5292, Japan

²⁾SOKENDAI (The Graduate University of Advanced Studies), Toki, Gifu 509-5292, Japan

³⁾Kinki University, Higashi-Osaka, Osaka 577-8502, Japan

⁴⁾Institute for Environmental Sciences, Obuchi, Rokkasho, Aomori 039-3212, Japan

(Received 7 October 2015 / Accepted 1 February 2016)

A deuterium plasma experiment is being planned at the Large Helical Device (LHD) at the National Institute for Fusion Sciences (NIFS). To delineate the regional background tritium concentration level before initiation of the experiment, we evaluated tritium concentrations in environmental water samples (river water, pond water, well water, tap water, and rainwater) collected at Tono area, Japan since 1982. Tritium concentrations in environmental water samples ranged widely from N.D. (below the instrumental detection limit of 0.27 Bq L^{-1}) to a maximum of 4.39 Bq L^{-1} . Tritium concentrations at 9 continuous monitoring locations over the 15 years ranged from N.D. to 1.36 Bq L^{-1} . This regional background concentration range will be used to evaluate environmental assessments after the initiation of the deuterium plasma experiment in LHD.

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Keywords: NIFS, LHD, tritium monitoring, environmental water sample, deuterium plasma experiment

DOI: 10.1585/pfr.11.1305032

Tritium (^3H) is a radioisotope of hydrogen ($T_{1/2}$: 12.3 years). Several natural sources of tritium exist. The majority of natural tritium is derived from nuclear reactions between secondary cosmic rays and nitrogen or oxygen atoms in the upper atmosphere [1]. The resulting tritium may be oxidized to form tritiated water [HTO], and HTO may migrate to the troposphere. Nuclear weapon testing in the atmosphere, conducted predominantly from the 1950s to the mid-1960s, released significant amounts of tritium into the environment. Although atmospheric tritium concentration rapidly increased because of the impact of released artificial tritium, it decreased gradually after this period, returning to almost natural background concentrations in Japan [2]. Since then, nuclear facilities such as nuclear reactors and nuclear fuel reprocessing plants have continued to release tritium into the environment. In addition, nuclear fusion reactors will use tritium as a fuel in the future. Although tritium fuel is burned in the core plasma and contained within the facility, small amounts of tritium may be released as gaseous waste exhaust products and wastewater effluents, which may be released into the surrounding environment [3].

The Large Helical Device (LHD), constructed by the National Institute for Fusion Science (NIFS) at Toki, Gifu Prefecture, is a heliotron-type plasma experiment device designed to investigate the extrapolatable plasma to the fusion reactor [4–6], and was started operation in 1998.

The deuterium plasma experiment (D experiment) is planned to higher performance plasma as the next project for 9 years. In the D experiments, a small amount of tritium will be produced by the $\text{D}(\text{d}, \text{p})\text{T}$ reaction that occurs from within the deuterium plasma [7]. The maximum annual amount of tritium production is currently 37 GBq in the first 6 years, and 55.5 GBq in the remaining 3 years. Over 95% of produced tritium is planned to be removed by using a tritium recovery system [8]. However, small amounts of tritium are expected to be released to the environment surrounding the NIFS site.

To precisely evaluate the environmental impact of tritium, it is important to understand the background concentration of tritium in the environment surrounding the NIFS site. Tritium concentrations in environmental water samples have been continuously monitored at Toki city, Tajimi city, and Mizunami city since 1982 before the construction of the NIFS facilities (for the experimental fusion program “R-project”) [9]. Tritium concentrations have also been analyzed from tap water collected at the Institute of Plasma Physics (IPP), Nagoya University, which is located 30 km southwest of the NIFS site. Part of the data of natural water samples has already been reported elsewhere [9, 10]. In addition, recent tritium concentrations in the atmosphere and plant samples have been already reported elsewhere [11, 12]. However, there are no recent reports on tritium concentration in environmental water samples in this area. In this paper, the tritium monitoring methodology and the

author's e-mail: tanaka.masahiro@nifs.ac.jp

Table 1 Chronology of tritium monitoring method for liquid scintillation counting.

	1982/8 -	1989/11 -	1992/8 -	1996/2 -	2005/5 -	2012/2 -
	20	8	17	10	9	9
Sampling points	A-1, A-2, A-3, A-4, B-1, B-2, B-3, C-1, C-2, D-1, D-2, D-3, F-1, F-2, F-3, H, K, X, Y, Z	A-1, A-3, B-1, B-3, C-1, F-1, F-2, Z	A-1, A-3, A-4, B-1, B-3, D-1, D-2, D-3, F-1, F-2, H, S, Z, W, X, Y, R	A-3, A-4, B-1, F-1, F-2, S, Z, X, Y, R	A-3, A-4, B-1, F-1, F-2, Z, X, Y, R	A-3, A-4, B-1, F-1, F-2, Z, X, Y, R
Sample volume (mL)	40	45	45	65	65	65
Total volume (mL)	100	100	100	130	130	130
Sample vial	Teflon	Teflon	Teflon	Polyvial	Polyvial	Polyvial
Liquid scintillator	Aquasol 2	Pico Flour-LLT	Pico Flour-LLT	Ultima Gold-LLT	Ultima Gold-LLT	Ultima Gold-LLT
Liquid scintillation counter	LSC-LB1 (Aloka, Japan)	LSC-LB1 (Aloka, Japan)	LSC-LB1 (Aloka, Japan)	LSC-LB3 (Aloka, Japan)	LSC-LB3 (Aloka, Japan)	LSC-LB5 (Aloka, Japan)
Background count rate (cps)	0.06	0.05	0.05	0.06	0.06	0.05
Counting efficiency (%)	13	12	12	15	15	18
Detection limit (Bq L ⁻¹)	0.53	0.46	0.46	0.28	0.28	0.27

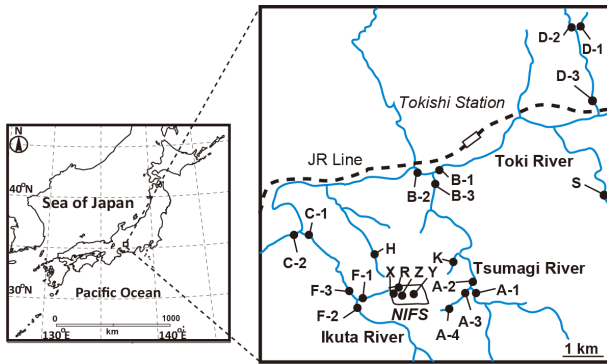


Fig. 1 Sampling location of environmental water at Tono area, Japan.

chronological data in environmental water sample by NIFS environmental monitoring program to understand the recent background tritium concentration level in this area.

Figure 1 shows the sampling locations in the Tono area. The watershed of the study area is dominated by the Toki river basin (1,010 km²). The geological framework of the Toki river basin consists of strata from mid-Paleozoic to Quaternary Cenozoic. The area contains a wide distribution of Nohi rhyolites and Ryoke granites that are covered by a thin layer of Seto Group stratigraphy [13, 14]. The average annual precipitation of this area is 1700 mm [9], which is similar to the Japanese average annual precipitation amount.

Table 1 shows the summary of the sampling and tritium measurement procedure conducted in this monitoring program. Most of water samples are collected from rivers, ponds except X, Y, Z, and R, where they are flow-controlled by NIFS, drain water, tap water of NIFS, sub-soil flow of the NIFS site, and rainwater. The S point is

well water. One to three liters of water samples were collected in washed polyethylene bottles. The sampling interval was conducted every 3 months (February, May, August, and November). The rainwater samples were collected using polyethylene bottles with a polyethylene funnel, every 3 months. The monitoring program was conducted at 20 sampling locations between August 1982 and November 1989. Thereafter, monitoring was conducted at 8 locations until May 1992 and at 17 locations from August 1992–November 1995 [10]. The water samples were collected at 10 locations from February 1996 to February 2005. Since May 2005, 9 locations are monitored. Water samples were also collected from 4 separate sampling points (A-1, B-3, C-1, H) as a backup for abnormal datasets in the main monitoring results. Each water samples about 200 mL was distilled after passing through a membrane filter (pore size, 0.45 μm). After distilling water samples, part of the sample was mixed with the same amount of a liquid scintillator, and counted by a low-background liquid scintillation counter for 1500 min (50 min × 15 repeat × 2 cycle).

Figure 2 show the change of tritium concentration in environmental water samples from August 1982 to November 2014. The all data ranged from N.D. (<0.27) to 4.39 Bq L⁻¹, with the highest value observed in November 1984 (F-2). Concentrations decreased over the observation period. The tritium concentrations in 1980s, 1990s, and after 2000, ranged from 0.51 to 4.39 Bq L⁻¹, N.D. (<0.28) to 2.65 Bq L⁻¹, and N.D. (<0.27) to 1.72 Bq L⁻¹, respectively. On its downstream side in Aichi Prefecture, the Toki River is referred to as the “Shonai River”. A similar trend has been reported in the Shonai River as well [15]. Tritium concentrations in rainwater collected since the 1960s was reported by the database of National Institute of Radiological Sciences [16] and MEXT [17]. Tritium concentrations in rainwater collected from Chiba ranged from 0.13

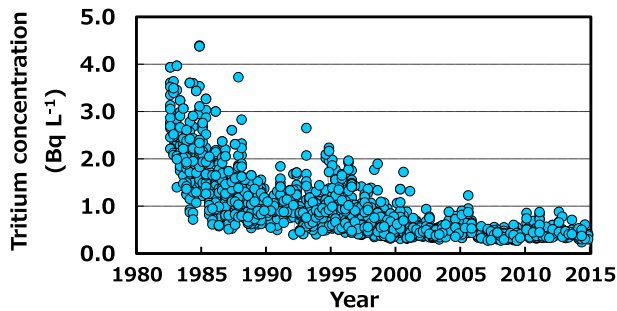


Fig. 2 Time series variation of environmental water samples collected at Tono area, Japan.

to 2.20 Bq L^{-1} , gradually decreasing in same period [16]. The data collected in this present study of environmental water samples was slightly higher than the reported value at Chiba. Tritium concentration in river water are mainly affected by local rainwater, groundwater, and runoff water. In general, tritium concentrations in groundwater are higher than that in local rainwater. Deposited rainwater into the ground and migrated into the water catchment area. Added soil water also migrated from the groundwater to the river. The residence time of rainwater reaching to the river depends on the conditions of water catchment area. Slightly higher tritium concentrations, in comparison with previous declines, are sometimes observed in the data trends (E.g. August 2000, August 2005) from this area. Although the reason behind the slightly higher tritium concentrations and wide concentration ranges in the natural water samples is not clear, the influence of surface runoff water and the residence time of water in the ground may be the candidates.

We estimated the recent background tritium concentration levels in this area. The annual mean value and standard deviation were used as the background tritium concentration levels [18]. From 1992, tritium monitoring was continued at A-3, A-4, B-1, F-1, F-2, Z, X, Y, and R. Data derived from 2000 was used for calculating the annual mean value to represent the recent data trends. However, multiple blank columns are present in the instrumental data, which are caused by “below detection limit” and “operation error”. Therefore, the annual mean value of tritium concentrations was calculated using “detection limit value” where no data existed. Recent tritium concentrations in environmental water collected at 9 continuous monitoring locations near the NIFS site and the annual average values are shown in Fig. 3. The annual average tritium concentrations in natural water from 2000 to 2014 (15 years) ranged from 0.27 to 0.75 Bq L^{-1} , with a mean value of $0.41 \pm 0.08 \text{ Bq L}^{-1}$. Sugihara [19] reported that the average tritium concentration in river water collected in Fukuoka city (Japan) in 2007–2008 was $0.57 \pm 0.12 \text{ Bq L}^{-1}$, which is similar to our reported value. The apparent half-life of annual average tritium concentrations is ap-

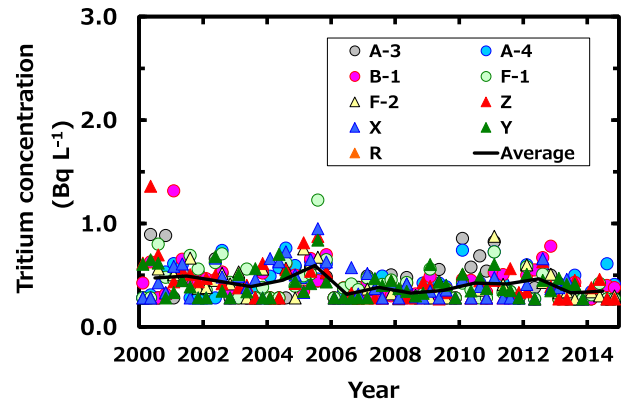


Fig. 3 Recent tritium concentration in environmental water and its annual average values collected at 9 points near the NIFS site.

proximately 32 years. Tritium concentrations in environmental waters, as illustrated in this study, may have wide concentration ranges depending upon sampling location, sampling timing, and weather conditions; in the 2000s, they varied yearly between N.D. (<0.27) and 1.36 Bq L^{-1} . This range will therefore be used as the recent concentration range. In the future, these values will be used as the standard level of environmental assessment during deuterium plasma experiments.

Acknowledgments

We acknowledge to Drs. H. Obayashi, S. Tanahashi, T. Uda (Emeritus Professors, National Institute for Fusion Science), Y. Sakuma, T. Kawano, and T. Saze (National Institute for Fusion Science) for their important support and discussion.

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