Monitoring of Tritium Concentration
by Simplified Active Sampler in a Fusion Test Facility*)

Masahiro TANAKA[1,2], Yukie YAMAMOTO[1], Chie IWATA[1] and Naofumi AKATA[1,2]

[1]*National Institute for Fusion Science, 322-6 Oroshi-cho, Toki 509-5292, Japan*
[2]*SOKENDAI (The Graduate University for Advanced Studies), 322-6 Oroshi-cho, Toki 509-5292, Japan*

From the viewpoints of radiation management and public acceptance, the monitoring of tritium concentration in the stack is one of the key issues for the deuterium plasma experiment in the Large Helical Device (LHD). Since 2012, an active tritium sampler was employed in the stack to monitor the background levels of tritium and discriminate between its chemical forms. However, the operation procedure became complicated, so we developed a simplified active tritium sampler to reduce the work after sampling. In this sampler system, there was no discrimination between tritium chemical forms, instead, all tritium chemical forms were collected in one process. To verify the performance of the simplified active tritium sampler, tritium concentration data was compared with the existing tritium active sampler and environmental tritium data at the NIFS site. The results revealed a correlation between these tritium data. Therefore, the simplified active tritium sampler was applied in the stack with a primary monitoring system for the deuterium plasma experiment in the LHD.

1. Introduction

The progress of nuclear fusion research reveals the importance of tritium. To produce high performance plasma, deuterium is used as the plasma operation gas. In the deuterium plasma experiment in the large fusion test device, tritium is produced by deuterium-deuterium reaction and removed from the vacuum vessel via a vacuum pumping system. Tritium in the exhaust gas is removed by a tritium recovery system, such as an exhaust detritiation system (EDS) or an air cleanup system. A small amount of tritium is then released into the environment via a stack. Since tritium is a radioactive material, which emits beta-rays and neutrinos, its management and control are one of the key issues in nuclear fusion facilities.

At the National Institute for Fusion Science (NIFS), the deuterium plasma experiment in the LHD is planned [1] and the instruments for radiation monitoring has been prepared. As for the environmental assessment, the distribution of environmental radiation at the NIFS site [2], including tritium concentration in environmental water samples (river water, pond water, well water, tap water, and rainwater) collected at the Tono area [3], free water tritium and organically bound tritium in pine needle samples collected at the NIFS site and nearby [4,5] and tritium concentrations of HTO, HT, and CH$_3$T in the atmosphere [4,6], are monitored prior to running the deuterium plasma experiment. However, from the viewpoint of radiation management in the control area, the following instruments for the management and control of tritium have been prepared: a portable-type tritium recovery apparatus, an expiratory water-trapping apparatus for internal tritium exposure measurement, a room gas monitor using an ionization chamber, an EDS for the LHD exhaust gas [7,8], a large-volume ionization chamber and active tritium samplers in the stack [9], etc. In tritium management, the monitoring of tritium concentration in the stack is crucial for the public acceptance. The regulation for tritium concentration in the stack varies between tritium chemical forms because the biological effect of tritium depends on its chemical forms. For example, the regulation in Japanese law is 5×10^{-3} Bq/cm^3 in the case of tritiated water vapor. However, the management standard at the NIFS, based on the agreement with local governments, is determined to be 2×10^{-4} Bq/cm^3, despite the various chemical forms. Since the tritium concentration is lower than the regulation in law, the accumulated active tritium sampler, which can measure a low tritium concentration, has been installed at the stack to provide the tritium background data before the deuterium plasma experiment in the LHD [9]. The active tritium sampler is able to discriminate tritium chemical forms. Consequently, post-processing data from the tritium sampler after sampling was labor-intensive. To reduce the load on the operator and the number of tritium water samples, we proposed a simplified active tritium sampler that did not discriminate chemical forms and evaluated
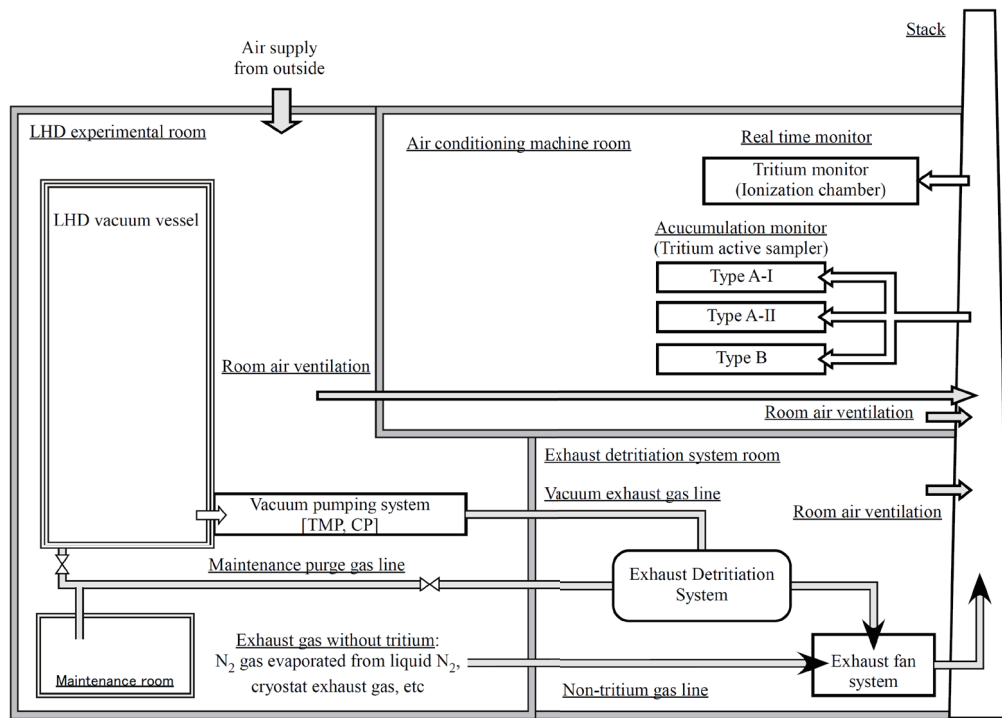
Fig. 1   Schematic of the LHD building, LHD exhaust gas line and active tritium samplersfor the stack.

the monitoring performance in the stack. In this article, the results of monitoring data by the simplified sampler are discussed, compared to the existing data in the stack and environmental data from the NIFS site.

2. Sampling System and Tritium Measurement

Figure 1 depicts a schematic of the LHD exhaust gas line with an integrated EDS and the surrounding building. The LHD has been in operation since 1998 to study plasma physics using hydrogen (H_2) gas [10]. Deuterium experiments have not, however, been conducted before 2016. Fresh air from outside is always supplied into the rooms for ventilation. Three tritium samplers were installed in the air-conditioning machine room of the LHD building and were connected to the sampling line in the stack. One sampler (Type A) is the simplified active tritium sampler and two samplers (Type A-I and Type A-II) are installed for system redundancy. The other tritium sampler (Type B) is for discriminating chemical forms.

A schematic of the simplified tritium sampler, Type A, is depicted in Fig. 2. The sampler has two paths for continuous sampling; the operational path switches after a given period of time. Air sampling is done at a flow rate of less than 2 L/min for one or two weeks. The total volume of sampled air was approximately $0.5 - 2\,m^3$. Type B also operates with the same sampling conditions.

In the air sampled by Type A, tritium, in the chemical forms of HT and CH_3T, was converted into HTO in an oxidation furnace with an alumina-supported palladium
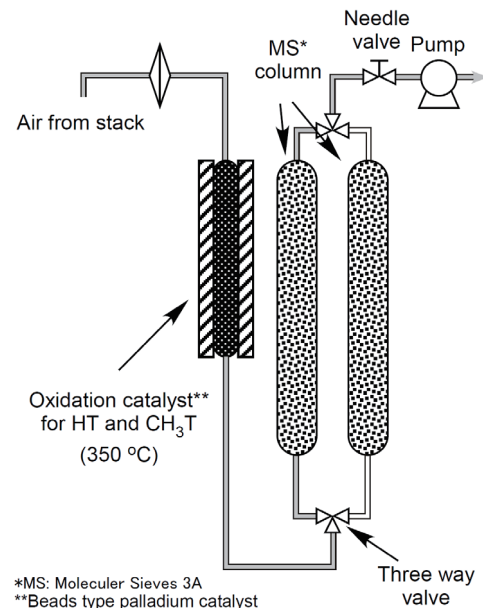

Fig. 2   Schematic of the simplified active tritium sampler (Type A).

catalyst (NE Chemcat, ND-101, Pd: 5 g/L, packed weight: 290 g, column shape: $\phi 40\,mm$ (O.D.) and 300 mmL) at 350°C. The oxidation performance of the catalyst has been evaluated in a previous article [9] and the conversion rate of methane oxidation reached unity at more than 200°C. The HTO formed was then collected together with HTO and H_2O in the air sampled in the absorbent column. Molecular sieves (Wako Pure Chemical Industries, LTD.) of 3A-type,

one-sixteenth inch pellet was used as the absorbent material. The dry molecular sieve was packed with approximately 550 g of the absorbent. Two absorbent columns were connected in series. As for the Type B system, after HTO in the sampled air was collected by the molecular-sieves column, tritium chemical forms of HT and CH_3T were oxidized by two kinds of catalyst at different temperatures. Then the HTO formed was collected by each molecular-sieve column. Thus, the Type B sampler can discriminate the chemical forms of HTO, HT, and CH_3T. More detailed specifications, as well as the flow diagram of the Type B sampler system, are described in a previous paper [9].

After sampling, the water collected in the molecular-sieve columns was desorbed at a temperature of 400°C and then recovered using a cold trap at approximately 2°C under a dry nitrogen (N_2) gas purge at a rate of 1 L/min for 3.5 h.

Tritium sampling by Type A-I was carried out from January 2015. Type A-II and Type B sampling were carried out from October 2016 and July 2012, respectively. In this study, the monitoring data until December 2016, before starting the deuterium plasma experiment, are demonstrated.

To measure the tritium activity, 20 mL of the recovered water from molecular sieves in Type A, and 20 mL of a liquid scintillator, Ultima-Gold LLT (Perkin Elmer Co. Ltd.), were mixed in a 20 mL Teflon vial. Background samples were prepared in 20 mL vials using tritium-free water. After leaving these samples for a few days in a low-background, liquid scintillation counter, LSC-LB-7 (Hitachi Co. Ltd.), the tritium activity was determined for a total of 1500 min per sample until November 2015 and 750 min per sample from December 2015.

3. Measurement Results and Discussion

The tritium monitoring results are depicted in Fig. 3. The data below the detection limit were omitted from the plots. The concentrations of HT and CH_3T measured by Type B did not vary with the season, as shown in Fig. 3 (c). However, it is well-known that the HTO concentration in the atmosphere varies with the variation of absolute humidity [4, 6, 11]. Therefore, the concentration of HTO measured by Type B increased in the summer. In the data of Type A-I, as illustrated in Fig. 3 (a), the tritium concentration increased in the summer. It is suggested that the chemical form of HTO increased with an increase of absolute humidity.

Figure 4 depicts the correlation between each tritium sampler. The sampling term is different between the samplers. Thus, comparable data in the same sampling term are extracted from the data in Fig. 3. Although the monitoring data of Type A-II are limited, they are in agreement with those of Type A-I. This suggests that there is not much
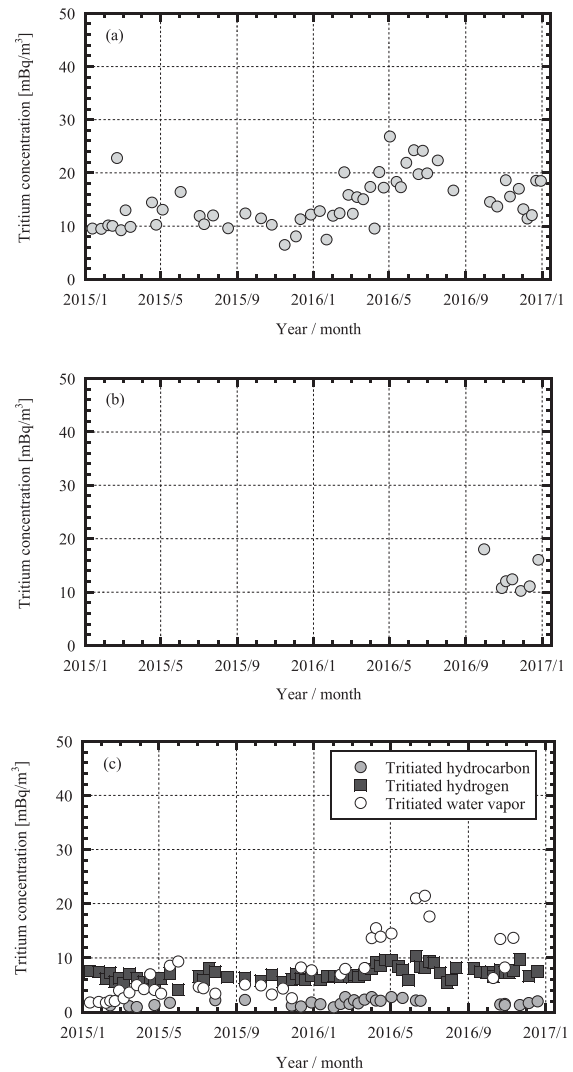

Fig. 3 Variations of tritium concentrations by different type samplers: (a) Type A-I, (b) Type A-II, (c) Type B (The data of Type B in 2015 is extractedfrom Ref. 9).

difference between same types of the sampler. Also, there is a correlation between Type A-I and Type B. Thus, the simplified active tritium sampler, Type A, can be applied for the monitoring system instead of Type B. The results of tritium monitoring would suggest a correlation between all tritium samplers, even though the range of tritium concentration is at an extremely low level.

The average tritium concentration, compared to each tritium sampler in this study and environmental data from the NIFS site, are summarized in Table 1. The levels of tritium concentrations in the stack were almost the same as the environmental tritium concentration (within the standard deviation) because ventilation by fresh air from outside was constantly supplied to the LHD building. The validity of the proposed, simplified tritium sampling system should be confirmed.

From the viewpoint of radiation control management, it is desirable that the radioactive waste material is reduced. The waste material of the simplified active tritium sampler,
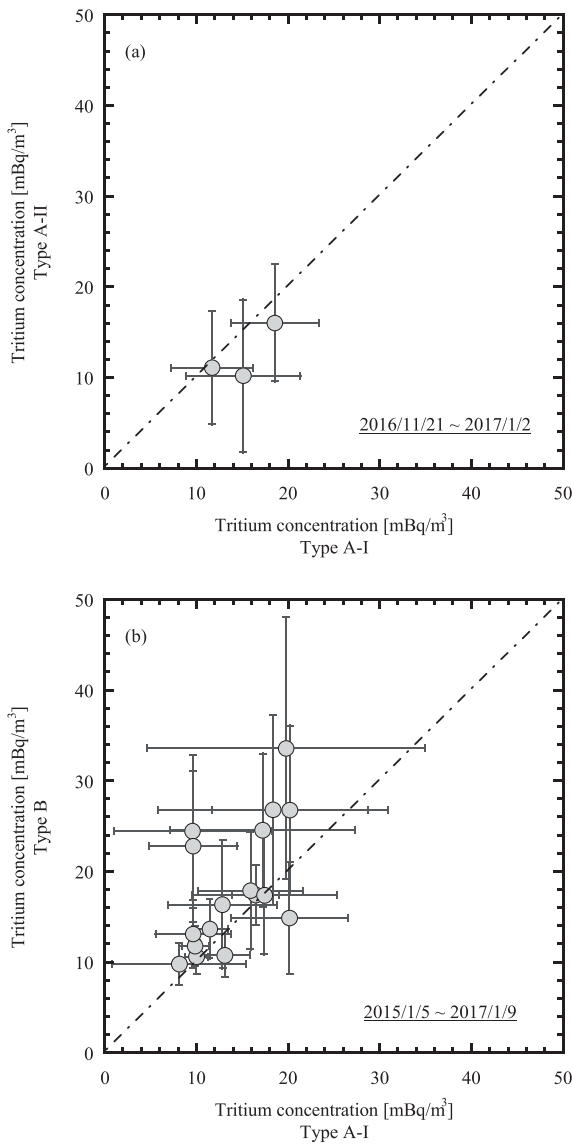
Table 1 Comparison of theaverage tritium concentration data among the active tritium samplerswith environmental tritium datafrom theNIFS site.

Location	Stack [This study]			Environment in NIFS site
Sampler type	Type A-I	Type A-II	Type B	Tritium sampler[12] for environment
Monitoring term	2015/1~ 2016/12	2016/10~ 2016/12	2015/1~2016/12	
Average tritium concentration [mBq/m³]	14.6±4.7	12.9±3.0	16.4±5.6	13.0±2.7

monitoring system in the fusion test facility.

4. Summary

A simplified active tritium sampler was proposed and developed for tritium monitoring in a fusion test facility. The proposed tritium sampler started operating from January 2015 before the deuterium plasma experiment in the LHD and monitored the air for validation in the stack. The range of the tritium concentration measured by the proposed sampling system was similar to that measured by the conventional sampling system and the environmental tritium level. It is suggested that the simplified tritium sampling system can replace the conventional tritium sampling system in the fusion test facility.

Acknowledgments

Fig. 4 Correlations of tritium monitoring data between each tritium sampler: (a) Type A-I and II, (b) Type A-I, and Type B.

Type A, is less than that of Type B, because Type A has only one column for absorbance and only one tritium sample is made. Also, the operator's work load is decreased using the simplified tritium sampler, Type A. Thus, the Type A sampler should be applied as the primary tritium

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