## §4. Observation of Environmental Tritiated Hydrogen and Methane Concentration at Toki Site

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The levels and the variations of environmental tritium at Toki site should be understood for the future experiments of LHD deuterium plasma discharges. Atmospheric tritium mainly consists of three different chemical forms HTO, HT and hydrocarbon like CH<sub>3</sub>T. Conventional technique of atmospheric tritium sampling is discriminate oxidization of tritiated species followed by collection of water with molecular sieve beds. For the monitoring of environmental tritium levels at Toki site, there is an accumulation of data acquired through observations of 6 years, 2004 - 2009. However, the concentration of tritiated methane in an environment is getting smaller and closing to the detection limit. Thus, to improve the detection limit for tritiated methane, the air sampling volume in winter shall be twice as much as last year for the increment of the collected amount of tritium. i) Sampling and radiation measurements

In 2010, the collected air volume was about 20 m<sup>3</sup> in summer and about 40 m<sup>3</sup> in winter at a rate of 500 ~ 2000 cm<sup>3</sup>/min once a month at the second flower of plasma diagnostic building in NIFS Toki site. The height of sampling point was about 6 m. The sampling time spent to 168 hours until 2006 and 300 ~ 650 hours from 2007. In July, the sampling term was 1 week because of the operation mistake. After collecting the air, water samples were recovered from the molecular sieves beds, which were regenerated at 400°C, passing through N<sub>2</sub> gas for 3.5 hours.

Radioactivity of tritium was counted by a low background liquid scintillation counter (LB-3 or LB-5, Aloka). The stocked water samples (65 g for HTO, 10 g for HT and CH<sub>3</sub>T) were mixed with the same amount of liquid scintillator (Ultima Gold LLT, Perkin Elmer). Twenty ml of vials were used for counting the HT and CH<sub>3</sub>T fractions while the HTO fractions were measured in 135 ml vials. Counting time was 1500 minutes for each sample, where measurements of 50 minutes were replicated for 15 times and the cycle was repeated twice.

The sampling data of January and February in 2011 was omitted because a count of the liquid scintillation sample of tritiated hydrogen and the amount of collected water vapor in air was abnormally low.

ii) Results of measurements

The measured data of tritiated hydrogen and methane concentrations from 2004 to 2010 are shown in Fig. 1. The concentration of tritiated hydrogen seems to increase in spring. The tritium in the stratosphere, which is produced by nuclear spallation reactions between cosmic rays and air, transports into the troposphere in spring. Then, the level of tritium concentration shall be increased and it calls "spring peak".

The atmospheric tritiated methane concentration level seems to vary widely before 2007. It is likely to show the variation of a short term sampling within 1 week. Then, after 2007, the variation of tritiated methane was suppressed and averaged because the sampling term became 1 month. However, in July of 2011, the tritiated methane concentration was about twice than other observed data in 2010. The sampling term in July was 1 week. Consequently, these results might indicate the variation of environmental tritium concentration with a short term tritium emission. On the other hands, the level of tritium concentration of HT and  $CH_3T$  in term of large volume sampling in 2010 is almost same as that in 2009. It is likely that a large volume sampling did not affect the observed tritium concentration. Therefore, it might be indicated that the level of tritiated methane concentration becomes low until few mBq/m<sup>3</sup> in recent years. This level of tritium concentration at Toki site is almost same level at Aomori.<sup>1)</sup>

From the viewpoint of the monitoring of the environmental tritium concentration, a large volume sampling is useful for the improvement of the detection limit. In the next fiscal year, we will try to increase the collection volume more than  $60 \text{ m}^3$  in winter.

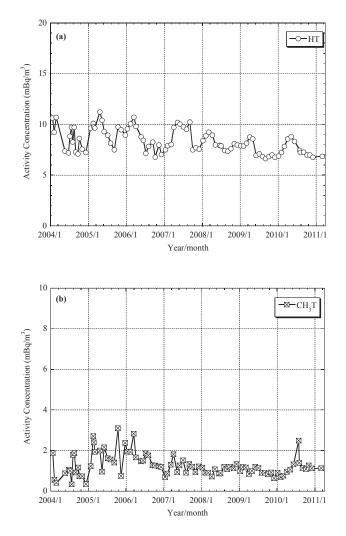


Fig. 1: Variation of atmospheric tritium concentration in 2004-2010 at Toki site; (a) HT, (b)  $CH_3T$ 

1) Igarashi, A., et al., Bulletin of Aomori Prefectural Nuclear Power Safety Center, 4, (2009), 71 [in Japanese].