

LEVELS OF ATMOSPHERIC TRITIUM IN THE SITE OF FUSION TEST FACILITY

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In the deuterium plasma experiment using Large Helical Device at the National Institute of Fusion Science (NIFS), a small amount of tritium is produced by the D-D fusion reaction. Then, a part of produced tritium is discharged into the environment via a stack. Thus, the atmospheric tritium in the site of NIFS has been monitored before starting the deuterium plasma experiment. The atmospheric tritium concentrations at NIFS were indicated to be background levels in Japan. To investigate the impact of tritium discharged from the stack, the correlation between the atmospheric tritium concentration and the tritium concentration observed in the stack was evaluated, and no significant correlation was found. In addition, the atmospheric tritium concentration at NIFS ranged within the background levels in Japan. Therefore, the impact of discharged tritium from the stack would be negligible in the environment at NIFS.

INTRODUCTION

At nuclear facilities such as nuclear power plants and spent fuel reprocessing facilities, tritium is continuously discharged into the environment⁽¹⁻⁷⁾. Therefore, to assess the impact of tritium in the environment, the tritium is monitored in the vicinity of the facilities. As one of the nuclear facilities, the nuclear fusion reactor will use hydrogen isotopes of deuterium (D) and tritium (T) as fuel. Therefore, the tritium monitoring around the nuclear fusion facilities would be an important issue.

In Japan, large fusion test devices such as JT-60U and Large Helical Device (LHD) have conducted the deuterium plasma experiment^(8, 9). In the deuterium plasma experiment, a small amount of tritium is produced in the vacuum vessel by the D-D fusion

reaction. Although the exhausted tritium is recovered by an exhaust detritiation system (EDS)^(10, 11) in the case of LHD, part of the tritium was discharged into the environment via the stack. As the result, the annual amount of discharged tritium for FY 2018-2020 ranged from 0.07 to 0.12 GBq⁽¹²⁻¹⁴⁾.

In this report, we focus on the tritium chemical forms of water vapor and hydrogen molecule, which are different biological effects, and discuss the impact of gaseous tritium discharge on the level of the atmospheric tritium in the environment at the NIFS site after the start of deuterium plasma experiments.

TRITIUM MONITORING IN THE NIFS SITE

Locations of the tritium monitoring and meteorological observation in the NIFS site

The NIFS site (35.326°E, 137.168°N) is located in central Japan, approximately 100 km inland from the Pacific Ocean and at a distance of 100 km or more from the nuclear power plants in Japan. The facility is situated on a hill, at an altitude of approximately 240 m above sea level.

The locations of the buildings and environmental monitoring stations in the NIFS site are shown in Figure 1. The Large Helical Device building, where LHD as a fusion test device is installed, is ventilated by the outside air intake and exhaust system. The ventilation through the building is discharged to the environment via the stack at the height of approximately 50 m from the

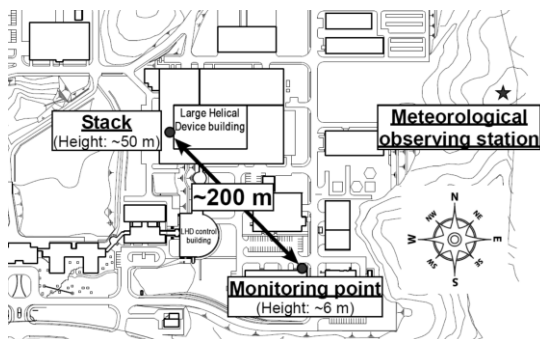


Figure 1. Locations of the stack, atmospheric tritium monitoring point and the meteorological observing station in NIFS site.

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ground. The tritium concentration in discharge gas is monitored by the active tritium sampler at the stack.

The meteorological observation station is installed at the top of the hill located 380 m away from the stack in the east direction. The height of the station is almost the same as the stack's discharge port. The monthly frequency of wind direction is summarized in Figure 2. The monthly mean wind speed tends to be stronger in winter and weaker in autumn. The range of variation is small, ranging from 2 to 2.5 m/s throughout the year. The wind direction in winter and spring is mainly from the west or northwest. In summer, the main wind direction becomes from the west and then varies to an east-west direction in autumn. Since the wind direction from northwest or west is relatively frequent and there is no suitable building in the east-west direction of the LHD building, the environmental tritium sampler was installed in the building 200 m away from the stack in the southwest direction.

Tritium monitoring instruments and tritium analysis

The active tritium sampler for the stack monitoring was designed for the discrimination of the chemical form of tritium without any combustible gas⁽¹⁵⁾ because the use of any combustible gas is restricted in the radiation-controlled area. The sampler mainly consists of three absorbent columns packed with molecular sieves 3A, two kinds of catalyst columns with different temperatures for the discriminating chemical forms of tritium, the water addition columns, a filter, a flow meter, a diaphragm pump. The discriminating chemical forms of tritium are water vapour, hydrogen, and hydrocarbons, but the data for water vapour and hydrogen are shown here. The sampling port in the stack was set at the height of 13–14 m from the floor level

because all ventilation lines were fed into the stack at the height of a few meters. In the stack monitoring, the collection period was set to two weeks throughout the year. The sampling flow rate was varied from 0.4 to 1.2 L/min depending on the season. As a result, the total collection volume was approximately 3 to 13 m³. After the completion of the collection, the absorbents in the active tritium sampler were regenerated, and the collected sample water for each chemical form of tritium was recovered. The detailed configuration of the system and the collection procedure are described in Tanaka *et al.*⁽¹⁵⁾. The collected sample water was mixed with a liquid scintillator (Ultima Gold LLT, PerkinElmer) in a 20 mL polyethylene vial at a ratio of 1:1. Then, they were measured by a low background liquid scintillation counter (LSC-LB7, Hitachi Co. Ltd.) in the radiation-controlled area for 750 min. The detection limit was approximately 1.2 Bq/L.

The active tritium sampler for the environment was designed for the discrimination of the chemical form of tritium using combustible gas⁽¹⁶⁾. The components of the sampler are almost the same as the sampler for the stack, however, the water addition columns are replaced by water electrolysis using tritium-free water and supply with methane gas from a cylinder. The sampling port was set at a height of 6 m from the ground. The chemical form discrimination was water vapour, hydrogen, and hydrocarbon, as in the stack. The collection period was set to about one month throughout the year. The sampling flow rate was varied from 0.5 to 2.0 L/min depending on the season. Then, the total collection volume was approximately 20 to 40 m³. As with the stack, the collected sample water was mixed with a liquid scintillator (Ultima Gold LLT, PerkinElmer) in a 1:1 ratio. A 130 mL polyethylene vial was used for the tritiated water vapour sample, and a 20 mL polyethylene vial was used for the tritiated hydrogen and hydrocarbon samples. Then, they were measured by a low background liquid scintillation counter (LSC-LB5 or LSC-LB7, Hitachi Co. Ltd.) in the non-radiation-controlled area for 1500 min. The detection limit was approximately 0.3 Bq/L for 130 mL vial and 1.0 Bq/L for 20 mL vial. The detailed system and the collection procedure are described in Uda *et al.*⁽¹⁶⁾.

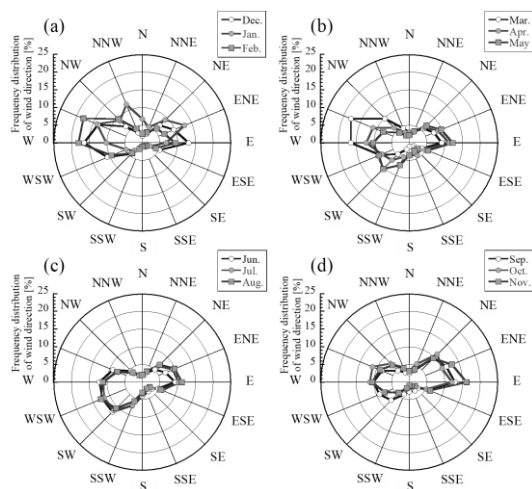


Figure 2. Annual wind direction diagram at NIFS site; (a) winter, (b) spring, (c) summer, (d) autumn.

RESULTS AND DISCUSSION

The tritium monitoring results at the stack and in the environment are shown in Figures 3 and 4, respectively. The deuterium plasma experiments were mainly carried out from October to January as shown by the arrows. Since the relative humidity of the air in the LHD building was controlled by the HAVC (Heating, Ventilation, and Air Conditioning) system, the absolute humidity in summer was less than that in the environment. The maximum tritium concentration in water vapour reached approximately 1.6 Bq/m³ during the LHD maintenance activity, which the workers

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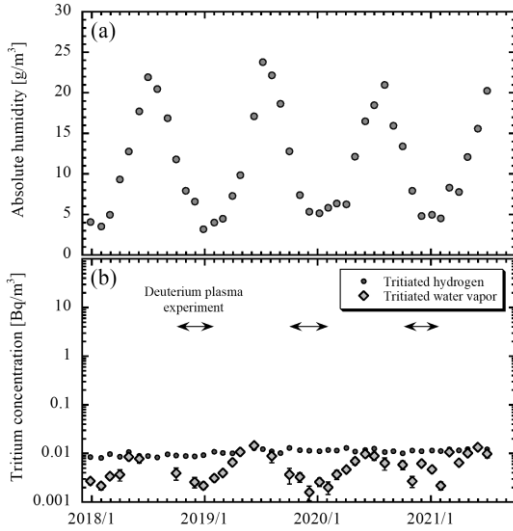


Figure 3. Variations of (a) absolute humidity and (b) tritium concentration in the stack from 2018 to 2021. The arrows indicate the period of deuterium plasma experiment.

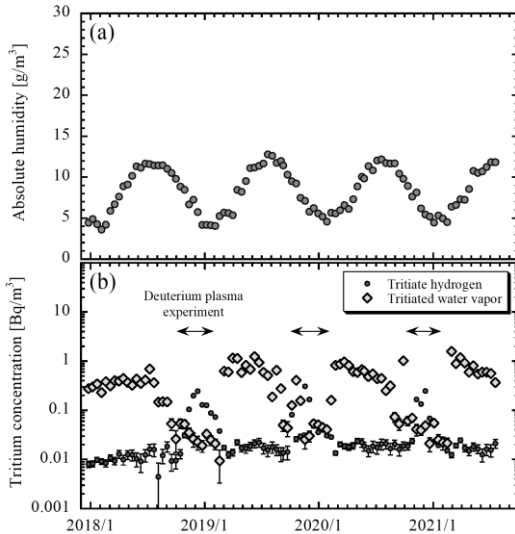


Figure 4. Variations of (a) absolute humidity and (b) tritium concentration in environment from 2018 to 2021. The arrows indicate the period of deuterium plasma experiment.

entered in the LHD vacuum vessel for the repair, the adjustment, the removal, and the installation of equipment, because tritiated water vapour was released from the LHD vacuum vessel. The minimum was approximately 0.01 Bq/m^3 which is the almost same as the maximum tritiated water vapour in the environment during the plasma experiment period.

On the other hand, tritiated hydrogen increased during the plasma experiment using hydrogen isotope gas, because the tritium chemical form in the exhaust gas during the plasma experiment was mainly hydrogen gas⁽¹⁷⁾. The maximum tritium concentration reached approximately 0.31 Bq/m^3 . Except for the period of the plasma experiment, the levels of tritiated hydrogen at the stack were almost the same as that in the environment. It would suggest that the tritiated hydrogen at the stack stands for the levels of atmospheric tritium because the LHD building is ventilated by the outside air.

The tritiated water vapour concentration in the environment depends on the variation of absolute humidity. Thus, the tritium concentration in water vapour becomes high in summer at NIFS as shown in Figure 4 (b). On the other hand, tritiated hydrogen is almost constant throughout the year. Although the tritium concentrations in water vapour and hydrogen at the stack changed with the situation of plasma operation and maintenance activity as mentioned above, it seems that there was no impact on the levels of atmospheric tritium in the environment by discharged tritium from the stack.

In addition, the correlation coefficient between the concentration of tritium in water vapour ($\text{Bq/L-H}_2\text{O}$) at the stack and in the environment is 0.3 and the p-value is below 0.05, indicating a weak correlation. It is supposed to be influenced by the correlation of absolute humidity variations and the situation of maintenance activity. For tritiated hydrogen, the correlation coefficient is -0.02 and the p-value is over 0.05. Thus, there would not be a correlation between the data of the stack and the environment.

The level of tritiated hydrogen in the environment seems to be increasing slightly since 2015⁽¹⁸⁾. In the last decade, the impact of anthropogenic tritium induced by atmospheric nuclear testing in the 1960s would become negligible, and then the natural variation of tritium produced by cosmic rays might be detected^(19, 20). According to Oulu neutron monitor data⁽²¹⁾, an increase in the neutron flux, which produces tritium by nuclear reactions between the cosmic rays and the atmospheric nuclei, has been observed since around 2015. Also, it could be a possible effect of tritium discharge from nuclear facilities around the world⁽²²⁾. To understand the production mechanism and the behaviour of atmospheric tritiated hydrogen related to cosmic rays such as solar activity cycle of 11-year, further observation of tritium and hydrogen (H_2) in the atmosphere may be necessary to obtain the additional data over the next decades.

CONCLUSION

The impact of tritium discharge on atmospheric tritium in the site of NIFS was evaluated for the deuterium plasma experiment using LHD as a large fusion device. To evaluate the environmental impact of

tritium discharged from the stack of the LHD building, the tritium concentration observed in the stack was compared with the atmospheric tritium concentration in the environment. As a result, no significant correlation was found between the tritium concentration variation in the stack and the atmospheric tritium concentration in the environment. The range of atmospheric tritium concentrations in the environment after the start of the deuterium plasma experiment was within the range of tritium concentrations before the start of the deuterium plasma experiment. Therefore, the impact of discharged tritium from the stack would be negligible in the environment at NIFS.

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REFERENCES

1. Fujita, H., Kokubun, Y., and Koarashi, J. *Environmental tritium in the vicinity of Tokai reprocessing plant*. J. Nucl. Sci. Technol. **44**, 1474-1480 (2007).
2. Vichot, L., Boyer, C., Boissieux, T., Losset, Y., and Pierrat, D. *Organically bound tritium (OBT) for various plants in the vicinity of a continuous atmospheric tritium release*. J. Environ. Radioact. **99**, 1636-1643 (2008).
3. Akata, N., Kakiuchi, H., Shima, N., Iyogi, T., Momoshima, N., Hisamatsu, S. *Tritium concentrations in the atmospheric environment at Rokkasho, Japan before the final testing of the spent nuclear fuel reprocessing plant*. J. Environ. Radioact. **102**, 837-842 (2011).
4. Kim, S.B., Bredlaw, M., and Korolevych, V.Y. *HTO and OBT activity concentrations in soil at the historical atmospheric HT release site (Chalk River Laboratories)*. J. Environ. Radioact. **103**, 34-40 (2012).
5. Connan, O., Hébert, D., Solier, L., Maro, D., Pellerin, G., Voiseux, C., Lamotte, M., and Laguionie, P. *Atmospheric tritium concentrations under influence of AREVA NC La Hague reprocessing plant (France) and background levels*. J. Environ. Radioact. **177**, 184-193 (2017).
6. Wu, X., Liu, Y., Kearfott, K., and Sun, X. *Evaluation of public dose from FHR tritium release with consideration of meteorological uncertainties*. Sci. Total Environ. **709**, 136085 (2020).
7. Hirao, S., and Kakiuchi, H. *Investigation of atmospheric tritiated water vapour level around the Fukushima Daiichi nuclear power plant*. Fusion Eng. Des. **171**, 112556 (2021).
8. Oikawa, A., Miya, N., Kodama, K., Umehara, T., Yamazaki, T., Masaki, K., Akiyama, I., Matsushita, K., and Hosogane, N. *Tritium experience in JT-60 DD plasma operation*. Fusion Sci. Technol. **41**, 612-616 (2002).
9. Osakabe, M., Isobe, M., Tanaka, M., Motojima, G., Tsumori, K., Yokoyama, M., Morisaki, T., and Takeiri, Y. *Preparation and commissioning for the LHD deuterium experiment*. IEEE Trans. Plasma Sci. **46**, 2324-2331 (2018).
10. Tanaka, M., Suzuki, N., Kato, H., Kondo, T., Yokosawa, M., Kawamata, T., Ikeda, M., Meguro, T., Tanaka, T., and Sono, K. *Design and commissioning of the exhaust detritiation system for the Large Helical Device*. Fusion Eng. Des. **127**, 275-283 (2018).
11. Tanaka, M., Suzuki, N., Kato, H., Chimura, H. *Performance of exhaust detritiation system for a fusion test device in the initial phase of the operation*. Fusion Eng. Des. **164**, 112172 (2021).
12. Annual Report for FY 2018 on the Activities of Radiation Safety in LHD deuterium plasma experiment. (2019), [In Japanese].
13. Annual Report for FY 2019 on the Activities of Radiation Safety in LHD deuterium plasma experiment. (2020), [In Japanese].
14. Annual Report for FY 2020 on the Activities of Radiation Safety in LHD deuterium plasma experiment. (2021), [In Japanese].
15. Tanaka, M., Kato, H., Yamamoto, Y., and Iwata, C. *Development of an active tritium sampler for discriminating chemical forms without the use of combustion gases in a fusion test facility*. Appl. Radiat. Isot. **125**, 53-59 (2017).
16. Uda, T., Sugiyama, T., Tanaka, M., Munakata, K., and Momoshima, N. *Developments of gaseous water, hydrogen and methane sampling system for environmental tritium monitoring*. Fusion Eng. Des. **81**, 1385-1390 (2006).
17. Tanaka, M., Suzuki, N., and Kato, H. *Exhaust behavior of tritium from the large helical device in the first deuterium plasma experiment*. J. Nucl. Sci. Technol. **57**, 1297-1306 (2020).
18. Tanaka, M., Akata, N., and Iwata, C. *Environmental Tritium around a Fusion Test Facility*. Radiat. Prot. Dosimetry **184**, 324-327 (2019).
19. Palcsu, L., Morgenstern, U., Sültenfuss, J., Koltai, G., László, E., Temovskii, M., Major, Z., Nagy, J.T., Papp, L., Varlam, C., Faurescu, I., Túri, M., Rinyu, L., Czuppon, G., Botyán, E., and Jull, A.J.T. *Modulation of cosmogenic tritium in meteoric precipitation by the 11-year cycle of solar magnetic field activity*. Sci. Rep. **8**, 12813 (2018).
20. László, E., Palcsu, L., and Leelössy, L. *Estimation of the solar-induced natural variability of the tritium concentration of precipitation in the Northern and Southern Hemisphere*. Atmos. Environ. **233**, 117605 (2020).
21. Oulu neutron monitor data. <http://cosmicrays.oulu.fi/>
22. Happell, J.D., Östlund, G., and Mason, A. *A history of atmospheric tritium gas (HT) 1950-2002*. Tellus, **56B**, 183-193 (2004).