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TRITIUM, HYDROGEN AND OXYGEN ISOTOPE COMPOSITIONS IN MONTHLY PRECIPITATION SAMPLES COLLECTED AT TOKI, JAPAN

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Monthly precipitation samples have been collected at Toki, Japan from November 2013 to March 2017. In this report, selected data were analyzed to identify the regional hydrogen and oxygen isotope compositions. Tritium (^{3}H) concentration in the precipitation ranged from 0.10 to 0.61 Bq L^{-1} and higher ^{3}H concentrations were observed in spring rather than in other seasons. This range was similar to values reported in Chiba City, Japan. ^{3}H concentration and the ratio d-excess, and δD values were roughly clustered according to each separate season. These regional hydrogen and oxygen isotope compositions will be used for environmental assessments of effects of the deuterium plasma experiments of the large fusion test device.

INTRODUCTION

Tritium (^{3}H) is a beta emitter radioisotope of hydrogen with a mean energy of 5.7 keV and half-life of 12.3 years[1,2]. Most natural ^{3}H is produced through the interaction of cosmic rays with ^{14}N and ^{16}O atoms in the upper atmosphere. On the other hand, artificial ^{3}H in the general environment has been produced by nuclear weapon testing and releases from nuclear facilities. Nuclear weapon testing in the atmosphere from the 1950s to mid-1960s released significant amounts of ^{3}H, and ^{3}H concentration in precipitation increased significantly[3]. Nuclear facilities such as nuclear power reactors and nuclear fuel reprocessing plants release ^{3}H during their normal operation. Elevated levels of ^{3}H concentration in atmospheric environment have been observed around nuclear facilities[4]. In the future, nuclear fusion reactors will have large ^{3}H inventories as fuel.

The Large Helical Device (LHD) constructed by the National Institute for Fusion Science (NIFS) at Toki City, Gifu Prefecture, is one of the biggest plasma experimental devices of the heliotron type[5]. At NIFS, deuterium plasma experiments will be conducted to investigate high-temperature plasma physics and the hydrogen isotope effect using the LHD. In the deuterium experiments, a small amount of ^{3}H will be produced by a fusion reaction. For the purpose of environmental assessment, ^{3}H concentrations in environmental samples (river water, pond water, well water and tap water) including precipitation were obtained every three months around the NIFS site[6]. In general, precipitation is monitored to evaluate the background ^{3}H level[7]. On the other hand, stable hydrogen and oxygen isotope compositions of precipitation are a useful tool to understand the physical processes which lead to ^{3}H formation and transport[8].

The aim of this study, therefore, was to clarify the background concentration of ^{3}H and stable hydrogen and oxygen isotope compositions in monthly precipitation samples collected at Toki City before undertaking the deuterium plasma experiments.

MATERIAL AND METHODS

Monthly precipitation samples were collected at NIFS in Toki City, Gifu Prefecture (35°19'N, 137°10'E), located as shown in Figure 1. Toki City is located approximately 30 km northeast of the Nagoya metropolitan area in central Japan. The average monthly precipitation was high in summer and low in winter, and average monthly temperature ranged from 9.2 to 33.7°C[9].

Precipitation samples have been collected from November 2013 to March 2017 by using a precipitation sampler (ST-1F, Suntechno, Japan) with a 10 L polyethylene container. After measuring sample weight, pH (B-211, Horiba, Japan) and electrical conductivity (EC) (E-771, Horiba, Japan), samples were distilled and

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enriched from 800 mL to 65 mL by a solid polymer electrolytic enrichment system (XZ001, De Nora Permelec Ltd., Japan). After distilling the ^{3}H -enriched sample water, 50 mL of sample water was mixed with the same volume of a liquid scintillation cocktail (Ultima Gold LLT, PerkinElmer, USA) in a 145 mL low diffusion polyethylene vial with inner Teflon coating (LDPE vial). ^{3}H radioactivity was measured using a low background liquid scintillation counter (LSC: LSC-LB5 or LSC-LB7, Hitachi, Japan) for 1,500 min. Counting efficiencies were determined by standard ^{3}H solution (SRM 4361C, NIST, USA). The minimum detection level (MDL) of LSC-LB5 and LSC-LB7 was approximately 0.31 Bq L^{-1}, therefore, the MDL of enriched sample water was approximately 0.04 Bq L^{-1}.

Stable isotope analysis was performed using both an isotope ratio mass spectrometer (Delta V Advantage, Thermo Fisher Scientific, USA) with a water equilibrium device (Nakano Electric Inc., Japan) and cavity ring-down spectroscopy isotopic water analysis (model L1102-i, Picarro Inc., USA) with a CTC Analytics autosampler (HTC-PAL, Leap Technologies, USA). Measurement precision was better than ±1.5‰ for δD and ±0.15‰ for δ^{18}O.

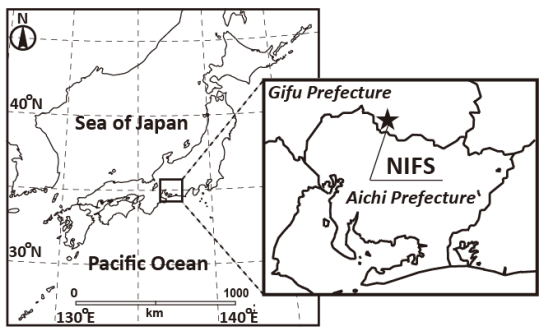

Figure 1 Location of sampling point

RESULTS AND DISCUSSION

Figure 2 shows precipitation data collected at Toki during the sampling period (November 2013 to March 2017): (A) pH, (B) EC, (C) monthly precipitation amount and (D) ^{3}H concentration. The pH ranged from 4.5 to 6.9, and EC ranged from 5 to 28 μS cm^{-1}. This result was comparable to the other reported value[13], and it seemed that the precipitation was not contaminated. Monthly precipitation ranged from 29 to 341 mm and was high in summer. Annual precipitations in 2014, 2015 and 2016 were 1515, 1435 and 1550 mm, respectively. The ^{3}H concentration in precipitation ranged from 0.10 to 0.61 Bq L^{-1} with a mean value of 0.32±0.12 Bq L^{-1}. Its seasonal trend was similar to the general background pattern which was high in spring and low in the other three seasons. ^{3}H concentration data in monthly precipitation in Japan have been already reported. The ^{3}H concentration in precipitation at Rokkasho Village, northern Japan during 2001 to 2006 ranged from 0.18 to 1.23 Bq L^{-1} with a mean value of 0.60±0.27 Bq L^{-1}[10]. A database gave data for Chiba City, Japan that ranged from 0.12 to 0.53 with a mean value of 0.30±0.10 Bq L^{-1} during the same period [11]. The present study results were slightly lower than those of Rokkasho, and similar to those of Chiba. Momoshima *et al.* [12] reported that ^{3}H concentration in precipitation in Japan depended on sampling latitude and was high in the north and low in the south. Akata *et al.* [10] reported that the peak concentration should be considered to be controlled by the air mass arrival from the Asian continent to the sampling site. The air mass transporting course would be the same for Toki and Chiba.

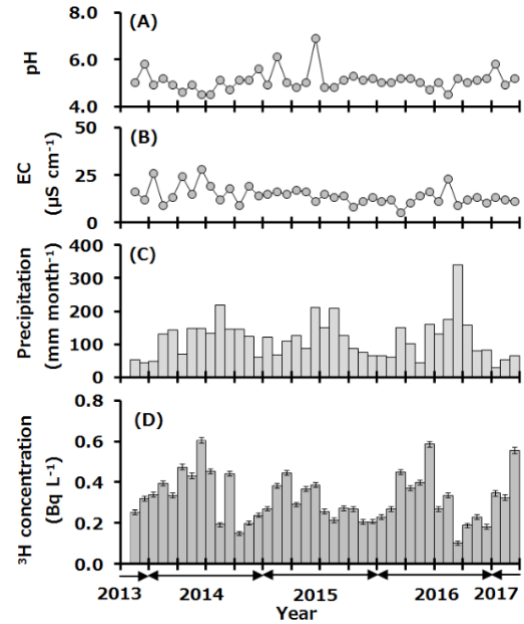

Figure 2 Data collected at Toki in precipitation: (A) pH, (B) EC, (C) monthly precipitation amount, and (D) ^{3}H concentration

Figure 3 shows the monthly mean ^{3}H concentration in precipitation at Toki during the indicated months from 2014 to 2016. Those values can be used as the monthly background value for calculating the contribution of ^{3}H from the deuterium plasma experiments.

Figure 4 shows the δD, δ^{18}O and d-excess in monthly precipitation at Toki. The δD values ranged from -103.62 to -20.77 ‰, and δ^{18}O values ranged from -15.14 to -3.92 ‰. There was no clear seasonal change. The d-excess values ranged from 0.1 to 29.7, but there was a clear seasonal trend of being high in winter and low in summer. In general, deuterium excess (d-excess) is used as tool for estimating the contribution of different

water vapor origin ("d-excess" = $\delta D - 8 \times \delta^{18}O$). In Japan, the northwestern monsoon from the Asian continent blows onto the Japanese archipelago. This monsoon carries dry air masses with water vapor evaporated rapidly from the Japan Sea. On the other hand, high-pressure systems develop in the Pacific Ocean in summer and bring air masses from the Pacific Ocean[14]. Therefore, it is important to determine the relationship between ^3H concentration and d-excess, and δD. The unit of ^3H concentration usually used is the tritium unit (TU) where 1 TU is defined as a particular isotope fraction equal to one ^3H atom per 10^{18} hydrogen atoms (1 TU = 0.118 Bq L^{-1}). Figure 5 shows the seasonal relationships between ^3H concentration (TU) and two quantities measured in the precipitation at Toki, (A) d-excess and (B) δD. Here, data groups were categorized for each season considering these data set and weather conditions[9] (spring, March-June; summer, July-September; fall, October and November; and winter, December-February), although general season categorization in Japan is every 3 months (spring, March-May; summer, June-August; fall, September and November; and winter, December-February). It is necessary to accumulate more data for reliable analysis. In the future, the authors plan to assess the ^3H released from the LHD that enters the environment as precipitation based on these background hydrogen and oxygen isotope properties.

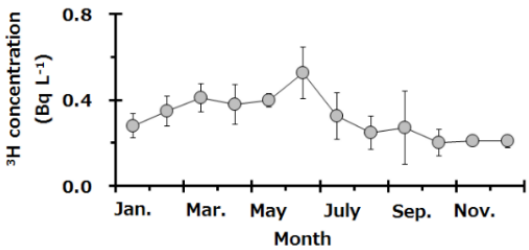

Figure 3 Monthly mean ^3H concentration in precipitation at Toki during the indicated months from 2014 to 2016

CONCLUSIONS

Hydrogen and oxygen isotope compositions in monthly precipitation have been observed since November 2013. From these observations, background ^3H concentration and δD and $\delta^{18}O$ composition and d-excess were reported. ^3H concentration in the precipitation ranged from 0.15 to 0.61 Bq L^{-1} and was high in spring and low in the other seasons. Although δD and $\delta^{18}O$ had no clear seasonal trend, d-excess did, and it was high in winter and low in summer. The ^3H concentration (TU) and the ratio d-excess, and δD were roughly clustered according to each separate season. These hydrogen and oxygen isotope compositions will be used for environmental assessments of the effects from deuterium plasma experiments carried out in the facility of NIFS.

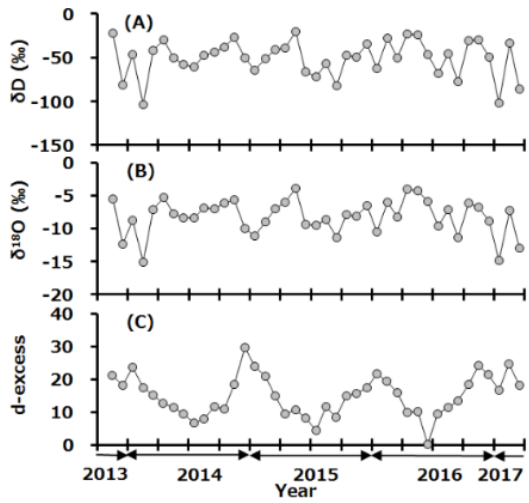

Figure 4 (A) δ D, (B) $\delta^{18}O$ and (C) d-excess variations in monthly precipitation at Toki

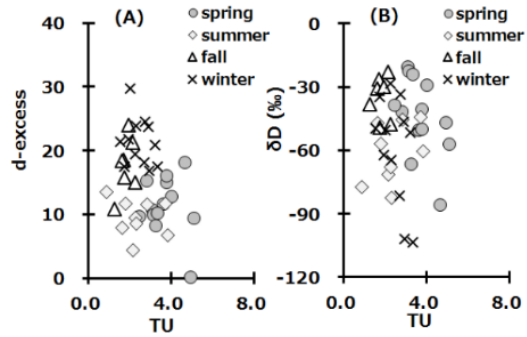

Figure 5 Seasonal relationships between TU and (A) d-excess and (B) δD in monthly precipitation at Toki

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