

Development of Field Estimation Technique and Improvement of Environmental Tritium Behavior Model^{*)}

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To estimate the environmental transfer of tritium into the atmosphere and to establish tritium analysis techniques, this study improves our previous compartment model and proposes new analytical technique based on the microwave heating method and a water-sample purification technique using ion resin. This article introduces a new conceptual approach for estimating environmental tritium behavior.

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1. Introduction

The Large Helical Device of the National Institute for Fusion Science (NIFS) started deuterium plasma experiments in 2017 [1]. The deuterium plasma experiments will assist our physical understanding of high-temperature plasma. However, to realize the fusion reactor, deuterium-tritium plasma experiments are necessary from both physics and fusion engineering perspectives. Such experiments were completed by the Joint European Torus and the Tokamak Fusion Test Reactor in the 1990s, and will be finished by the International Thermonuclear Experimental Reactor (ITER) [2–4] in the near future. Tritium is a radioactive material that emits beta-rays with a maximum energy of 18.6 keV. To ensure the safety of fusion facilities, methods that evaluate tritium transfer in the environment are vital. When gaseous tritium (HT) and/or tritiated water (HTO) are/is released from a facility into the atmosphere, HT and HTO disperse and are deposited on soil, plants, terrestrial water or seawater surfaces, and are eventually transferred to the body interior [5]. In particular, re-

leased HT is oxidized to HTO by bacteria in the soil. The dose coefficient of HTO is 10,000 times higher than that of HT [6]. HTO in the soil, water, and air is transferred to plants, where it becomes free water tritium (FWT); moreover, organically bound tritium (OBT) formed by photosynthesis accumulates in the plant and is slowly discharged from it (see Fig. 1).

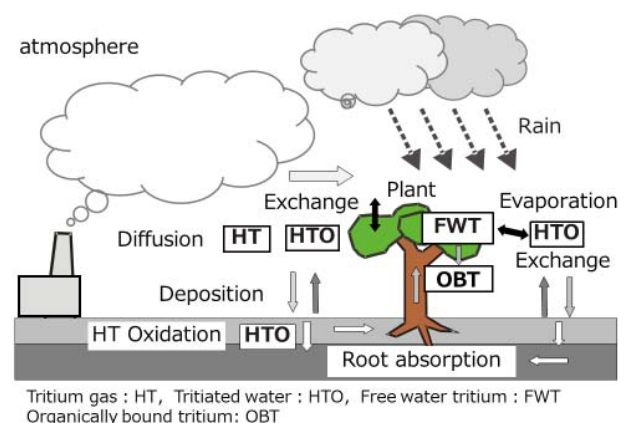


Fig. 1 Tritium transfer in terrestrial environments.

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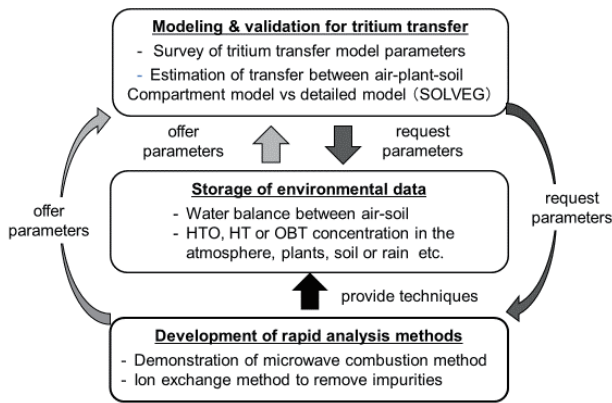


Fig. 2 Outline of this research project.

The present study aims to develop a tritium analysis technique and to improve the tritium transfer model based on the simple compartment model, establish practical parameters, and hence estimate tritium transfer in the environment. The models are developed under normal operation conditions of the facilities in previous studies. Here we describe the improvements to the model, and develop an analytical technique for obtaining the parameters. To obtain several environmental sample data, we also establish methods for rapidly analyzing liquid scintillation measurements of plant and water samples. An outline of this research project is shown in Fig. 2.

2. Tritium Behavior Model

The structure of the developed model is shown in Fig. 3. This model is composed of an air-soil-plant system. Tritium in plants is divided into FWT and OBT. The OBT component is subdivided into fast and slow components, each containing exchangeable OBT (Ex-OBT) and non-exchangeable OBT (Nx-OBT) according to recent knowledge [7]. The source terms of this model are the HT and HTO concentrations in air. Although tritiated methane (CH_3T) can also be released from nuclear facilities such as fuel reprocessing plants, we focused on HT and HTO because the solubility (and hence the plant availability) of methane in water is low. The transfer coefficient of each pathway would be estimated by using previous studies and from experimental or field data obtained in this study. To validate the developed model, the results would be compared with those of SOLVEG-II [7].

3. Methods for Estimating Soil-Water Transfer

Soil water was collected by a lysimeter system with glass fiber wicks (Fig. 4). This system is inserted perpendicularly to the soil-water flow in the soil profile at a specific depth beneath undisturbed soil. As no vacuum is applied, saturated flow conditions are established at the upper boundary of the sampler, enabling water to perco-

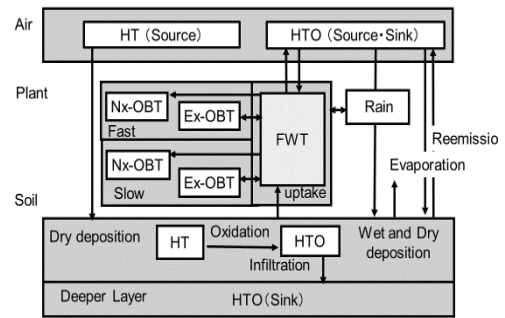


Fig. 3 Developed compartment model for tritium transfer in the terrestrial environment.

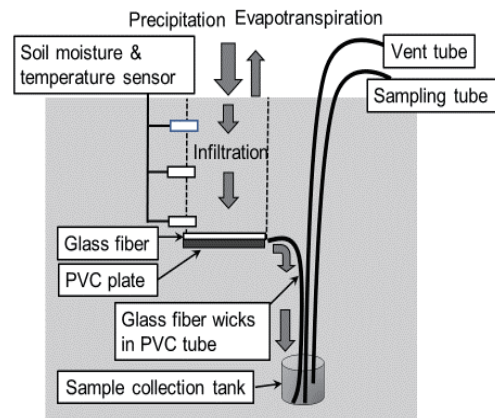


Fig. 4 Schematic of the system for sampling soil water.

late into the system. After removing the percolate with a pump, the volume of soil water and tritium concentration in the soil water can be measured. This sampling system is simple, non-destructive, and convenient for assessing soil-water transfer. The system can acquire continuous monitoring data, and provides information on travel time. The system, constructed from a 0.5 cm-thick polyvinylchloride (PVC) plate, provides a sampling area of 0.25 m² which is covered with glass fiber. In the present study, the sampler plate was set at 20 cm below the soil surface. The sampler was connected to a 4-L high-density polyethylene container by flexible PVC tubing. From the data collected by the lysimeter system, we attempted to estimate the soil-water transfer.

4. Environmental Tritium Analysis by the Microwave Heating Method

Conventionally, FWT and OBT are analyzed by the liquid scintillation counting (LSC) method after freeze-drying and combusting the sample. However, the pretreatments for FWT and OBT analysis are complicated and time-consuming, requiring several weeks. To reduce the time and effort of pretreating the plant samples, we propose a microwave heating technique prior to the FWT and OBT analysis. We first tried recovering the water from

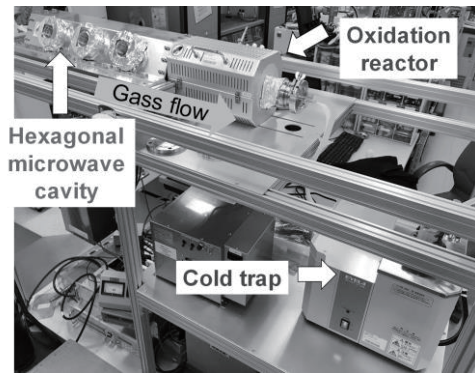


Fig. 5 Photograph of the hexagonal microwave heating system.

fresh pine needle samples by a multimode-type large-volume microwave cavity with 2.45-GHz magnetron-type microwave generators, whose basic parameters had been determined in advance. The cavity collects the complete moisture content of the sample. After the microwave heating experiments, the dried pine needle samples were re-dried in an oven dryer for calculating the FWT recovery yield. The recovery yield exceeded 97% under the imposed experimental conditions (microwave output: 1.0 kW, time: 30 min, N_2 flow: 2 L min^{-1}). This result indicates that sample drying and water recovery by microwave irradiation is more time-efficient than the conventional method [8]. We then designed and constructed a new pretreatment microwave heating system composed of the multimode-type hexagonal microwave cavity, 2.45-GHz magnetron-type microwave generators, an oxidation reactor packed with a palladium catalyst, and a combusted water recovery cold-trap (see Fig. 5).

We preliminarily tested the combustion of plant powder. The dried plant sample was completely combusted after approximately 2 hr. In future, we will optimize the hexagonal microwave heating system for OBT measurement using a semiconductor microwave generator.

5. New Analytical Methods for Pretreatment and Tritium Enrichment

5.1 Pretreatment of water samples for liquid scintillation measurements

To suppress chemical quenching in the LSC method, we must remove the impurities from the sample water. The conventional pretreatment method is distillation of the sample water, which is another time-intensive process. Sample water for tritium LSC measurements has recently been purified through ion-exchange resins, as shown in Fig. 6 [9]. Toward an easy and effective LSC pretreatment, we thus investigated the performance of anion- and cation-exchange resins and tritium columns produced by Eichrom. We checked the absorption spectrum in the UV range and the electrical conductivity of the water sam-

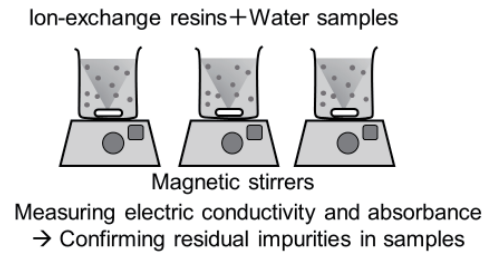


Fig. 6 Simple water-purification method.

ples to confirm the water quality (organic and ionic-species contents in the recovered water).

The results confirmed that all impurities were removed from the simulated sample. As the next step, we will attempt to purify more contaminated simulated samples.

5.2 Development of a compact electrolytic tritium-enrichment cell for small water samples

The tritium concentration in environmental water samples such as rivers, rain, and seawater is too low to measure directly by the LSC method. Although an electrolytic tritium-enrichment system for environmental samples is commercially available [10], it requires a large-volume water sample and limits the electrolysis current. For this reason, we developed a compact electrolytic tritium-enrichment cell for small-volume water samples. The glass cell holds approximately 300 mL of water sample, and the designed volume after enrichment is 12 mL. The electrolysis employs three electrode plates (Ni-Fe-Ni) and proceeds at 2.4 g/hour. In this system, the detection limit of the electrolytic enrichment of a 60-mL water sample equaled that of direct measurement of a 50-mL water sample in a 100 mL vial. Enrichment by this system will be effective if sample volumes of 100 mL or higher can be obtained. In a 100-mL water sample, the expected detection limit is 0.15 Bq/L for 1,000 min monitoring by LSC measurement.

6. Summary

In future, the environmental behavior of tritium will be effectively evaluated by the simple compartment model, and the model parameters will be obtainable by the rapid and simple techniques developed in our research project.

Acknowledgment

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