4.21 The Evaluation of Uptake of Tritiated Methane to the Plants

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ABSTRACT

The experiment of the exposure of tritiated methane (CH_3T) to plants was carried. Plants were put in the airtight container that CH_3T gas was added. The amount of light that it irradiated plants was changed, and the amounts of photosynthesis as the metabolic change activities of the plants were adjusted, and processes of assimilation to the plant of CH_3T were evaluated under a constant temperature. The leaves in the container and the inside gas were collected in every interval, and tritium concentrations in the samples were determined.

It is observed that CH_3T concentration in the container decreased under both the light and dark conditions. On changing from dark to light condition, there was no change in the decreasing tendency of the CH_3T concentration. These show that tritiated methane would be taken in the plants without the photosynthesis process, but through the oxidation by microorganism on plants.

KEY WORDS: tritium, methane, plant, photosynthesis

1. INTRODUCTION

Most tritium (T) released from a variety of nuclear facilities is generally in the molecular form (tritiated hydrogen gas - HT or T_2 - referred to here as HT) or the oxide form (tritiated water - HTO, DTO or T_2O - referred to here as HTO). Some tritium may also be released in various organic chemical forms, mostly as tritiated methane gas ($CH_{4-x}T_x$- referred to here as CH_3T) [1]. Tritiated methane will be released from not only nuclear detonation but also nuclear fusion reactors and burial sites of radioactive wastes including tritium [2]. Among them the migration behavior of discharged HTO and HT are rather well known including uptake by plants, but knowledge of discharged organic forms of tritium is lacking [2]. In the environment, the chemical form of the released tritium may change. Tritium released as HT may be converted to HTO with subsequent transformation to OBT, some released as HTO may become bound as OBT in biota. Photosynthesis is the mechanism by which most of the primary production occurs in ecosystems. Plant incorporates carbon dioxide into organic molecules through photosynthesis. Stomata in the leaf can open or close to control the movement of carbon dioxide in and water vapor out of the leaf. Tritiated water vapor is easily taken in the body in plants through their stomata. The possibility of direct uptake of atmospheric tritiated methane by plants was suggested by Belot [3] and Amano [4], though methane gas is known as inert in photosynthesis of plant.

It is important in public health to evaluate the intake of tritiated compound to the plant in steady state and toward an unusual release from the nuclear power facilities. It is necessary to clear the intake mechanism of tritiated methane to the plant in detail.

2. EXPERIMENTAL

2.1. Overview of the experiment

The experiment of the exposure of tritiated methane to plants was planned. Plants were put in the airtight glove box chamber that tritiated methane gas was added. Several potted plants having different photosynthetic processes were exposed to tritiated methane in the enclosed chamber over three days. The exposure chamber was constructed of acrylic boards. The volume of the chamber was *ca.* 140 L. Over the upper part of the chamber, there are nuorescent lamps. The plants were exposed to a total of 98 MBq of tritium in the form of methane mixed with the chamber. The methane carrier gas was 630 ppm. Photosynthesis is the light reactions, and the intensity depends upon the light intensity. The amount of light irradiated plants was changed, and the amounts of photosynthesis as the metabolic change activities of the plants were controlled, and processes of assimilation to the plant of tritiated methane were evaluated under a constant temperature. The temperature in the chamber was within 26 +/- 1 °C during the whole experiment. The intensity of light was shown in Table 1. Exposure began in the dark condition. Tritiated methane gas in a glass break-seal ampoule was released into distilled groundwater which has no tritium, to prevent the exposure of tritiated water and soluble tritiated molecules to plants in the chamber. The potted plants exposed to tritiated methane included young tomato (*Lycopersicon esculentum* MILL.) as a C3 plant, Indian corn (*Zed mays* L) as C4 plant, and aloe (*Aloe arborescens* Mill.) as a CAM plant. Each pot was covered with a plastic film to prevent the reaction of methane gas with the potted soil [5]. In addition, the surfaces of covered film and the inside wall of chamber were sterilized by 70% ethanol solution to prevent the microbial interaction with methane gas. Only the leaves and branches were exposed to tritiated methane.

Table 1. Light intensity in the chamber during the experiment.

Time (h)	Intensity (lx)
0 - 4	0
4 - 4.5	500
4.5 - 8	30,000
8 - 12	20,000
12 - 20	10,000
20 -	0

2.2. Sample treatment

The leaves in the chamber and the inside gas were collected in every interval, and tritium concentrations in the samples were determined. The tritiated methane in the air was collected periodically in 5mL-glass vials and sealed with septum caps in the glove box chamber. After CH_3T exposure, the collected air was introduced into cylinder of the ionization chamber (Ohkura Electric), and Concentrations of CH_3T in the chamber were determined.

The leaves were collected periodically in 50mL-glass vials and sealed with caps in the glove box chamber. After three days of exposure, sealed leaf samples were added to distilled water with no tritium, to extract the tissue free water tritium in the exposed leaves. The extracted water was added to a liquid scintillator, Pico Fluor LLT and radioactivity measured using a liquid scintillation counter, Packard 2250 LSC. The tritiated water vapor in the chamber was collected by adsorption on molecular sieves (MS-5A) in 20-mL glass vials. Then, they were added to distilled water with no tritium, to dilute the adsorbed tritiated water vapor, and radioactivity in supernatant fraction measured with Packard 2250 LSC.

3. RESULTS AND DISCUSSION

3.1. Tritium concentration in the chamber

Fig. 1 shows accumulation of tritium in the form of CH_3T with elapsed time after the initiation of CH_3T exposure. Amount of CH_3T in the chamber was showed arithmeric decrease at the beginning of the CH_3T exposure in spite of dark condition. Photosynthesis process does not work in the dark condition. The decrease in amount of CH_3T in the dark condition means that photosynthesis process did not involved in metabolism of methane gas.

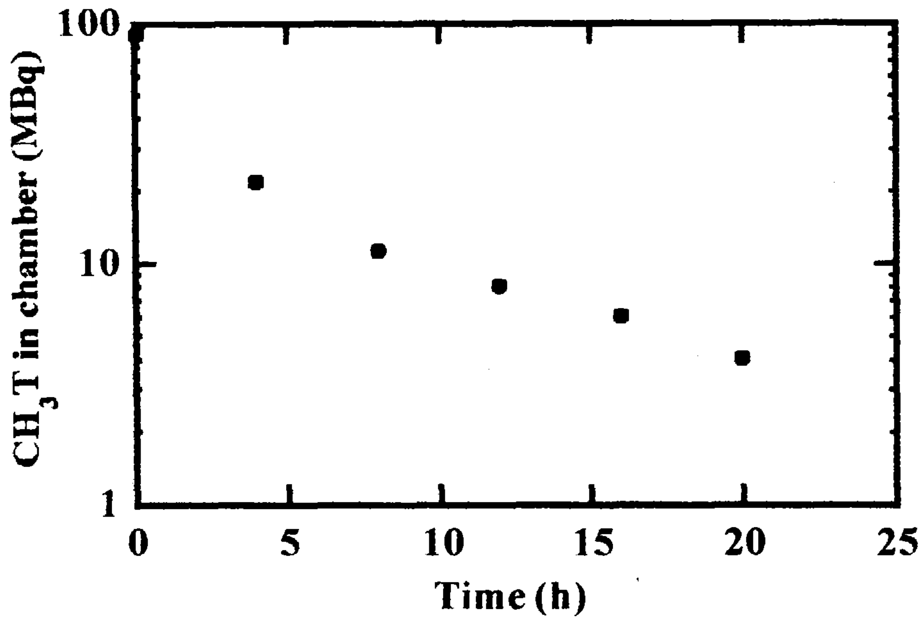

Fig. 1. Time course of tritiated methane (CH_3T) in the experimental chamber.
(The plots were average in duplicate.)

The dew condensed on the inside wall of the chamber was observed. Relative humidity was anticipated to be saturated in the chamber at the end of CH_3T exposure, because of the water transpired from plants in the chamber. Fig. 2 shows accumulation of tritium in the form of HTO in water vapor with elapsed time after the initiation of CH_3T exposure. The drastic increase of HTO at 4h from the start of exposure was meant to be resulted in the oxidation of CH_3T, and strongly suggests the microbial oxidation of methane gas. The temporal decrease of HTO concentration at 8h from the start of exposure was observed. The decrease was assumed to be resulted in the transpiration. High intensity of light (30000 lx) between 4.5h and 8h promoted the transpiration from the plant body, and water vapor with low tritium concentration was released in the chamber. After 12h from the start of CH_3T exposure, most tritiated methane was oxidized to tritiated water, and tritium concentration in water vapor became steady value.

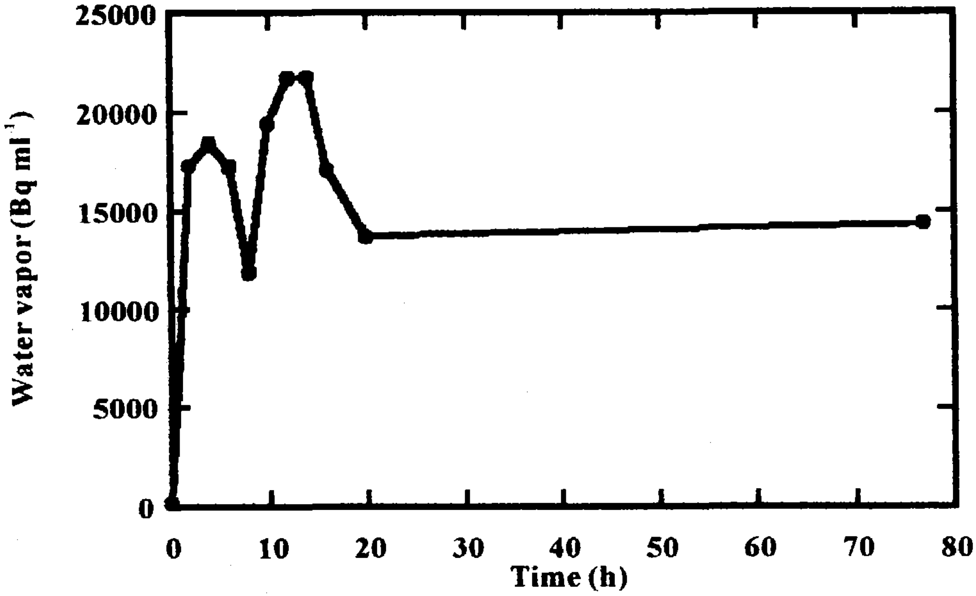

Fig. 2. Changes of HTO concentrations in the concentration in the experimental chamber. (The plots were average in duplicate.)

3.2. Tritium concentration in vegetation

Tritium was detected in the exposed leaves of C3 and C4 plants in the tissue free water. Fig. 3 shows accumulation of tritium in the form of FWT in several potted plants with elapsed time after the initiation of CH_3T exposure. FWT concentration in leaves of the tomato which are C3 plants gradually increased. FWT concentrations in leaves of the Indian corn which is a C4 plant was almost the same level as the C3 plants at 0-20h, but FWT in the aloe which is a CAM plant stayed at a very low level during the whole of CH_3T exposure, because of the short duration of the exposure of light by which photosynthesis activity proceeded. After 20h, FWT concentrations in leaves of tomato as C3 plant less increased compared with the Indian corn as C4 plant. There seems to exist potential mechanisms for transfer of tritium from tritiated methane in air to HTO in plant leaves. Some microorganisms living in the surface of plant leaves may convert some amounts of CH_3T to HTO. Tritiated methane may be oxidized on the leaf surface by methane oxidizing bacteria to tritiated water, which could transform into leaf cells where they could be readily metabolized and/or involve tritium exchange with cell water. Tritium was consider to be transferred into the leaves as water vapor after conversion of CH_3T to HTO.

Low FWT/HTO ratio (< 0.01) was observed in each plant. These values were quite low compared with the reported value [6]. High humidity may account for the low transpiration rates. A concentration of carbon dioxide in the chamber was increased to over 8000 ppm at the end of CH_3T exposure. The low ratios arose from suppress the transpiration through the stomata owing to large amounts of CO_2 and high humidity in the chamber. However, the HTO concentrations in the chamber were reflected on the FWT in vegetation [7-9].

This suggests that the transfer of tritiated methane to plants is general phenomena in C3 and C4 plant not dependent on the difference of the photosynthetic through conversion of CH_3T to HTO. CAM plant closed its stomata in daytime, so tritium did not enter into aloe.

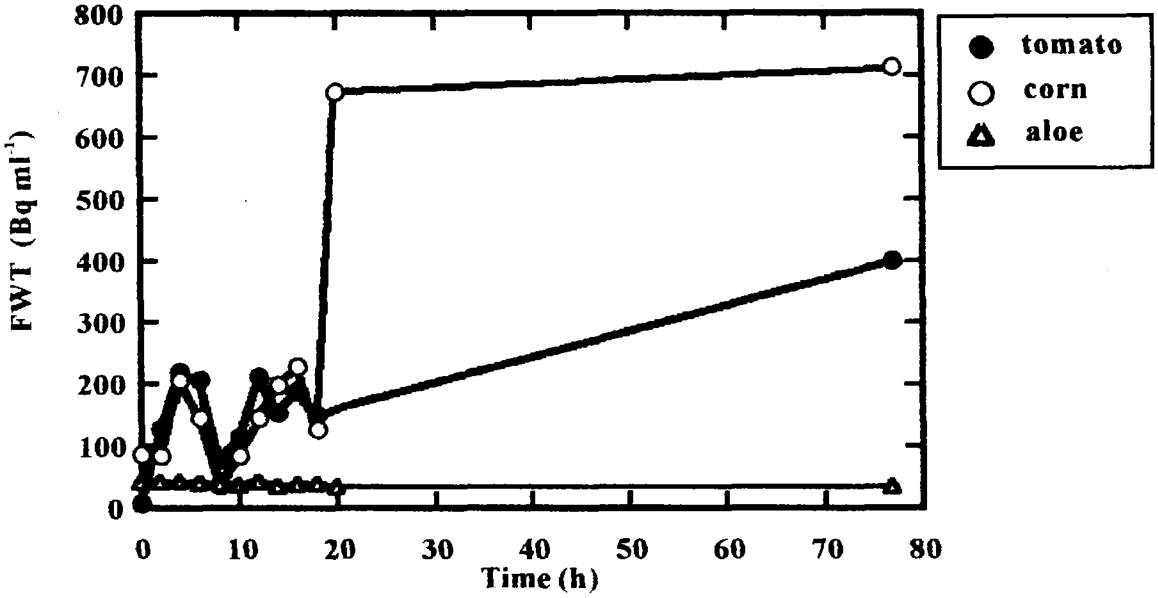

Fig. 3. Changes of FWT concentrations in vegetations (tomato, Indian corns, aloes) in the experimental chamber.
(The plots were average in duplicate.)

4. CONCLUSIONS

Tritium was detected in the plant leaves which are exposed to tritiated methane. CH_3T was rapidly oxidized into HTO in the dark condition. That is indicated that photosynthesis could have low or no influence on the oxidation of CH_3T. CH_3T was thought to be oxidized through the microbial processes on the plant surface in this experiment. The mechanism of accumulation tritiated methane in plant is not dependent on the photosynthesis.

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