Journal of Environmental Radioactivity 101 (2010) 177–184

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Migration of ^{14}C in the paddy soil-to-rice plant system after ^{14}C-acetic acid breakdown by microorganisms below the plow layer

Shinichi Ogiyama*, Hiroshi Takeda, Nobuyoshi Ishii, Shigeo Uchida

Office of Biospheric Assessment for Waste Disposal, National Institute of Radiological Sciences, 4-9-1 Anagawa, Inage-ku, Chiba 263-8555, Japan

ARTICLE INFO

Article history:
Received 14 July 2009
Received in revised form
8 October 2009
Accepted 14 October 2009
Available online 5 November 2009

Keywords:
Geological disposal
Organic carbon
Rice
Root-uptake
Transuranic waste

ABSTRACT

Migration of ^{14}C derived from ^{14}C-acetic acid was examined by using soils sampled from paddies in four administrative areas in Japan (Aomori, Yamanashi, Ehime and Okinawa) and rice plant in a tracer experiment to understand the fate of ^{14}C in the paddy soil-to-rice plant system. The loss of ^{14}C radioactivity levels derived from ^{14}C-acetic acid was caused by soil microorganism breakdown. A part of the ^{14}C fixation to soil was caused by microbial assimilation into the fatty acid fraction. ^{14}C moved upward via two different types of ^{14}C dynamics in soil: quick movement upward; and constant but slow movement upward. ^{14}C was highly assimilated into the plant panicle and that was caused by the root-uptake and the transfer of ^{14}C. Migration of ^{14}C derived from ^{14}C-acetic acid relied heavily upon changes of chemical forms and characteristics of ^{14}C-compound as caused by microorganisms in soil.

1. Introduction

Radiocarbon (^{14}C, $t_{1/2} = 5.73 \times 10^3$ yrs) is one of the main nuclides in transuranic (TRU) waste which is generated during the operation of radioactive waste reprocessing systems in reprocessing facilities. The geological disposal of TRU waste is planned to avoid radiation exposure to the general public. The concept of the safety assessment for its geological disposal mainly consists of three independent items: artificial barriers, natural barriers, and the biosphere. The biosphere is defined as the part of the earth containing living organisms and in which an ecosystem exists, and it is the last link in a chain of models that assess movement of various nuclides in the environment. The safety assessment scenarios for geological disposal of radioactive waste are described based on migration of radioactive nuclides flowing into the biosphere, including agro-ecosystems, through natural barriers and aquifers with upward movement of groundwater.

For public health safety, it is necessary to clarify pathways on how radioactive nuclides reach the biosphere and its human inhabitants. The main pathway for elemental intakes by humans is through food ingestion after the elements are absorbed by edible plants. In many cases, data from the technical report series 364 by IAEA have been used for models that assess transfer of various nuclides to edible plants (International Atomic Energy Agency, 1994). However, little is known about ^{14}C movement in the environment and ^{14}C transfer to edible plants via roots from geological disposal of TRU waste despite its uptake in the primary food sources of living organisms.

Organic carbon in TRU waste is mainly present as carboxylic acids, low-molecular alcohols and aldehydes (Kaneko et al., 2003). Among carboxylic acids, acetic acid and formic acid are the dominant species. However, carbon compounds easily change their chemical structure and, hence, their characteristics in the environment. Studies have reported that a large amount of the ^{14}C derived from acetic acid is released to the atmosphere in some gaseous form produced by microorganism breakdown in flooded paddy soils (Ishii et al., 2009), and major gases caused by ^{14}C-acetate degradation in wetlands are methane gas (CH_4) and CO_2 (Ström et al., 2003). Thus, to understand the fate of ^{14}C for environmental assessment of TRU waste disposal, it is important to monitor the fate of ^{14}C associated not only with water movement but also with gas transformations.

Since there is an astonishing richness of microorganisms on plant roots (Großkopf et al., 1998), carbon behavior in the soil and plant rhizosphere is also thought to be marked by changes of its chemical structure and characteristics. Based on observations of H_2 and several carbon component products, Conrad and Klose (1999) reported that the metabolic reactions such as methanogenesis were predominantly due to microorganisms living on the root surface. A major carbon source for plants has been generally thought to be

* Corresponding author. Tel.: +81 43 206 4606; fax: +81 43 206 4601.
E-mail address: ogiyama@nirs.go.jp (S. Ogiyama).

0265-931X/$ – see front matter © 2009 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jenvrad.2009.10.004

CO_2 from air, which is assimilated by photosynthesis. However, it is also necessary for safety assessment to clarify transfer of carbon from underground and carbon absorption by plant roots associated with microorganisms. Carbon is present in chemically changeable compounds in the environment which are both available and non-available forms for plant absorption; therefore, carbon from the plow layer associated with microorganisms as well as carbon from air is very important for carbon assimilation for edible plants in the context of geological disposal of TRU waste.

Because of the high consumption of rice in many Asian countries as a staple in the diet, particular attention should be paid to rice plants. Plant physiology provides very useful information to understand carbon transfer to a rice plant through its roots. Studies have reported that carbon moves to rice plant shoots in gaseous forms such as CO_2 via the roots (Higuchi, 1982; Higuchi et al., 1984). The transfers of gases from the root to the atmosphere via the plant have been described by diffusion (Lee et al., 1981). Such gases generally go up through the aerenchyma system of the rice plant and are emitted to the atmosphere through the stomata of the lower part of the leaf sheath. Although useful information has been obtained in various field studies, creation of the ^{14}C migration scenario for the paddy soil-to-rice plant system is still difficult because paddy fields are a very complex artificial wetland ecosystem. Against these conditions, elucidation of ^{14}C migration in soil and root-uptake of ^{14}C by the rice plant are among the most useful ways to understand the hazards of radiation exposure to human health through food ingestion for the scenario related to assessment of geological disposal of TRU waste.

A culture experiment made by the authors (Ogiyama et al., 2008) has shown that the amounts of ^{14}C-acetic acid absorbed by rice plant through the roots were very small. A large amount of $^{14}CO_2$ gas produced by ^{14}C-acetic acid breakdown was released from the rooted medium to the atmosphere. The $^{14}CO_2$ gas would be taken up by plants through the roots, and some fractions of $^{14}CO_2$ gas were assimilated into the shoots by photosynthesis. However, these results were obtained from hydroponics or sand culture experiments which are not enough to discuss ^{14}C migration in the paddy soil-to-rice plant system as related to geological disposal of TRU waste. In order to obtain further fundamental knowledge for discussion of geological disposal of TRU waste, the present study examined migration of ^{14}C, including ^{14}C-acetic acid degradation in soil, and ^{14}C plant uptake in the paddy soil-to-rice plant system after ^{14}C-acetic acid breakdown by soil microorganisms below the plow layer using soils collected from actual paddy fields.

2. Materials and methods

2.1. Soil samples

Soils were sampled in paddy fields of four administrative areas (prefectures) in Japan (Aomori, Yamanashi, Ehime and Okinawa). The soils were classified as Gley Lowland Soil, Gray Lowland Soil, or Lowland Paddy Soil according to the Cultivated Soil Classification Committee in Japan (National Institute for Agro-Environmental Sciences, 1996). Several physical and chemical properties of the soils (pH (H_2O), total carbon and nitrogen contents, bulk density (BD), and maximum water holding capacity (MWHC)) were determined. The pH (H_2O) was measured with a glass electrode pH meter using a sample to water ratio of 1–2.5. Total carbon and nitrogen contents were measured with a CHNS-analyzer (Eurovector, Euro EA3000, Milan, Italy) using 10 mg soil samples. Soil cores (100 mL) were collected using a coring instrument and weighed after drying them in an oven for the determination of the soil BD. Soil samples were also weighed before and after being saturated with water for determination of the MWHC.

2.2. ^{14}C-radioactivities in soil as obtained in the incubation experiment and by fatty acid analysis

For the incubation experiment, the fresh paddy soil sample of 600 g (approximately 400 mL; 330–460 g on a dry weight basis) was placed in a 500 mL plastic beaker. Acetic acid labeled with ^{14}C (1,2–^{14}C) was added to the beaker to give a radioactivity of 185 kBq (0.055 μmol of ^{14}C-acetic acid). The soil samples were well mixed using a glass rod, and then were covered with water for the entire experimental period to simulate a paddy field. The soil samples were incubated for 30 d at 25 °C. During this period, soil portions were collected from each beaker at day 0, 7, 12, 19 and 30 after the ^{14}C-acetic acid addition to examine changes of ^{14}C radioactivity levels in soil. The soil sample of 0.5 g on a dry weight basis was combusted, and resultant $^{14}CO_2$ was trapped in liquid carbon absorber (Carbo-sorb E, PerkinElmer) by using a sample oxidizer (Packard, Model 307). The absorber solution was mixed with a scintillation cocktail (Permafluor E^+, PerkinElmer). ^{14}C radioactivity in the solution was then determined using a liquid scintillation counter (PerkinElmer 3100 TR). Sample treatments were done in duplicate.

Experimental glass columns (50 cm in height, 50 mm in diameter) containing the paddy soil spiked with ^{14}C-acetic acid of 185 kBq were prepared. The columns were filled with the soil to a depth of approximately 25 cm, (410 ± 50 g of dry soil), and one 2-month-old rice seedling (*Oryza sativa* L. cv Koshihikari) was cultivated in each column for 158 d in a condition with natural light (10 k Lx), 28 °C, and relative humidity of 55%. After the rice plant was harvested, the soil in the column was mixed for uniformity, and the ^{14}C radioactivity in soil fatty acid fraction was determined to clarify the distribution of ^{14}C derived from acetic acid in soil microorganisms. Four experimental steps were required to cleave the fatty acids from lipids: saponification for destruction of the microorganism cell membrane, methylation of free fatty acid, organic extraction of methylated fatty acid, and base wash for elimination of impurities (Sasser, 2001). A fresh soil sample (5 g on a dry weight basis) was collected from each column and placed in a 50 mL test tube. Saponification was done at 100 °C for 30 min in the test tube using 6 mL of a solution of 45 g NaOH, 150 mL C_3HO, and 150 mL water. Then, methylation was done at 80 °C for 10 min by adding 9 mL of a solution of 325 mL 6 N HCl and 275 mL CH_3OH. For organic extraction of fatty acid, methyl ether (6 mL of a 1:1 mixture of hexane and methyl tert-butyl

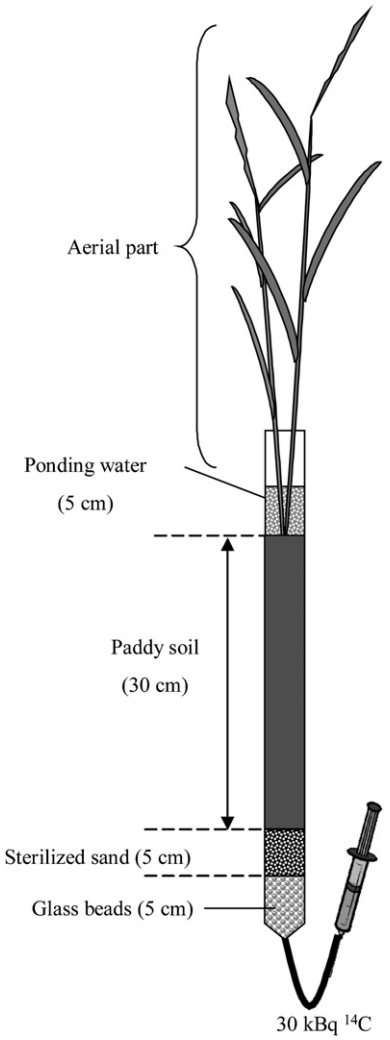

Fig. 1. Schematic drawing of the set-up used for culture experiment. ^{14}C-acetic acid solution was injected into the column bottom with a syringe.