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Soil-to-Rice Transfer of ^{99}Tc in Paddy Soils Contaminated in Two Different Ways

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1. Introduction

Rice is one of the most important food crops in the world. All isotopes of technetium (Tc) are radioactive, and the environmentally most important one is ^{99}Tc because of its very long half-life ($2.1x10^5$ years) and relatively high ^{235}U-fission yield [1,2]. Accordingly, it is one of the critical radionuclides in an environmental impact assessment for radioactive waste disposal. A significant amount of ^{99}Tc can be released into the atmosphere in a severe reactor accident as was shown in the Chernobyl accident [3]. It is a pure better emitter and thus internal exposure via food consumption may be a primary contributor to the ^{99}Tc radiation dose to humans [1]. Paddy rice fields can be contaminated with ^{99}Tc in various ways. In the present study, greenhouse experiments were conducted to investigate the transfer of ^{99}Tc from four paddy soils contaminated in two different ways. One was to simulate plowing the topsoil after a pre-transplanting deposition of ^{99}Tc, whereas the other was to simulate a ^{99}Tc deposition onto the surface water shortly after transplanting.

2. Materials and Methods

Experimental soils were collected in four paddy fields located around the Gyeongju nuclear site. The soils were carried to the laboratory and air-dried. Table I summarizes the major physicochemical properties of the dried experimental soils. A total of 20 lysimeters were prepared with 30.4 kg of dry soil contained in each. They were randomly placed in a greenhouse and irrigated 4 d before transplanting. For each pre-transplanting deposition lysimeter, 20 kg of dry soil and a radioactive premix (0.4 kg of dry soil and 30 ml of ^{99}Tc solution (247 kBq ml^{-1})) was thoroughly mixed using a mixing machine 23 d before transplanting (23DBT) and put into a lysimeter already containing 10 kg of dry soil. For each post-transplanting deposition lysimeter, 25 ml of the same solution was evenly applied onto the water surface using a micropipette 1 d after transplanting (1DAT). The surface water was first sampled 5 d and 3 h after the water contamination for the depositions at 23DBT and 1DAT, respectively. Thereafter, samplings were made seven times at an interval of 4-28 d for both.

Plants were harvested for the aerial parts 132 d after transplanting. The harvested plants were divided into the ears and straws. After being dried, the grains were separated from the ears and the chaffs were removed from the grains to obtain brown rice. The dried brown rice and straw samples were powdered using a mortar and mixer. The powder samples of brown rice were treated with ammonium hydroxide [4] and incinerated for 12 h in a muffle furnace. Total beta counting was performed for the ash samples of brown rice and the powder samples of straws.

Soil-to-rice transfer of ^{99}Tc was quantified with the values of TF_{ag} (aggregated transfer factor, m^2 kg^{-1}-dry), which were calculated as follows [1];

$$TF_{ag} = \frac{\text{Plant concentration of } ^{99}\text{Tc (Bq kg}^{-1}\text{-dry)}}{\text{Deposition density of } ^{99}\text{Tc (Bq m}^{-2}\text{)}} \quad (1)$$

Table I: Major Properties of the Experimental Soils

Soil	pH (1:5)	OM (%)	Clay (%)	Texture
Daebon	5.5	3.7	11.0	Loam
Gugil	5.1	4.2	15.4	Silt loam
Eupchon	5.6	3.0	26.9	Loam
Sangra	5.1	4.9	25.2	Loam

OM : Organic matter.

3. Results and Discussions

2.1 TF_{ag} Vaues

Table II shows the TF_{ag} values of ^{99}Tc for brown rice and straws in four paddy soils contaminated in two different ways. The straw values were markedly higher than the brown-rice values. This implies that the mobility of ^{99}Tc from the vegetative part to the seeds is very low. The TF_{ag} values from the 1DAT deposition were generally higher than those from the 23DBT deposition. This can be largely attributed to an increased contact of the root surface with the radionuclide following the former, as compared with the latter. The two soils having the lowest TF_{ag} values for the 23 DBT deposition had the highest values for the 1DAT deposition. One important reason for this may be that the ^{99}Tc from the 1DAT deposition may have migrated down through the aerobic soil layer much more slowly in the two soils than in the other soils mainly due to the higher clay contents. The transfer of ^{99}Tc from soil to rice plants following the 23DBT deposition can also be quantified with the traditional transfer factor, which is defined as the ratio of the plant concentration to the soil concentration [1].

Table II: TF_{ag} values of ^{99}Tc for rice plants following two different ways of soil contamination

Soil	TF_{ag} value (m^2 kg^{-1}-dry)			
	Brown rice		Rice straw	
	23DBT	1DAT	23DBT	1DAT
Daebon	6.2E−06	5.2E−06	4.4E−03	6.9E−03
Gugil	1.1E−05	1.6E−05	5.7E−03	9.6E−03
Eupcheon	2.3E−06	7.3E−05	2.3E−03	3.7E−02
Sangra	3.6E−06	7.0E−05	2.8E−03	4.1E−02
GM	4.8E−06	2.6E−05	3.6E−03	1.8E−02

GM: Geometric mean.

2.2 Concentrations of ^{99}Tc in surface water

Figs. 1 and 2 show the evolutions of the ^{99}Tc concentrations in the surface water following the depositions at 23DBT and 1DAT, respectively.

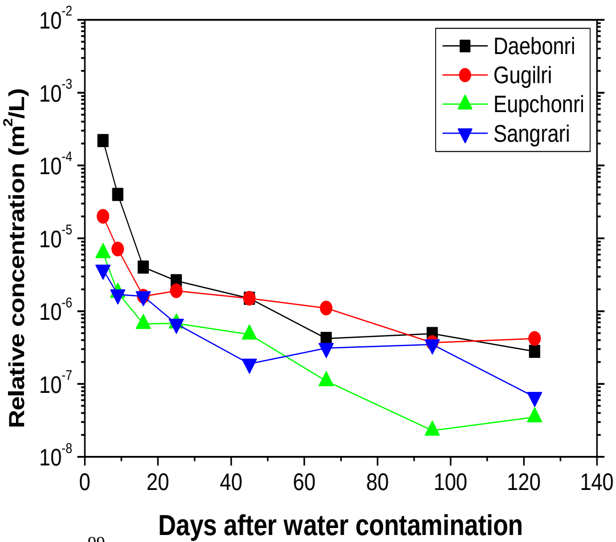

Fig. 1. ^{99}Tc concentrations in surface water following the deposition at 23DBT.

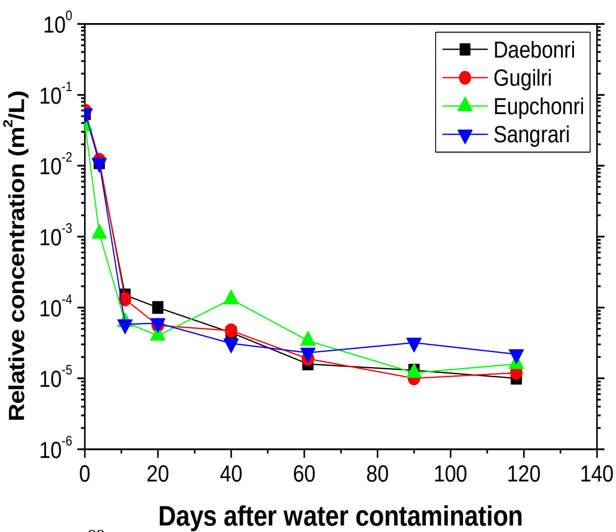

Fig. 2. ^{99}Tc concentrations in surface water following the deposition at 1DAT.

The concentrations of ^{99}Tc following the deposition at 1DAT (Fig. 2) were markedly higher than those for 23DBT (Fig. 1) over the whole period, indicating the possibility of a significantly higher uptake of ^{99}Tc via the plant base following the 1DAT deposition [5]. This anticipated higher plant-base uptake may be partly responsible for the difference in the TF_{ag} value between the depositions at 23DBT and 1DAT. Rapid decreases in the ^{99}Tc concentration occurred for the first about 10 or 15 d after the water contamination. This may be mainly attributable to a decrease in the solubility of ^{99}Tc due to reduction processes under anaerobic soil conditions.

4. Conclusions

Soil-to-rice transfer of ^{99}Tc in paddy soils was experimentally investigated for two different scenarios of the paddy-field contamination. It was demonstrated that a post-transplanting deposition of ^{99}Tc onto the surface water can lead to a much higher transfer than a pre-transplanting deposition followed by plowing. The surface-water concentrations of ^{99}Tc following the post-transplanting deposition was markedly higher than those following the pre-transplanting deposition, possibly indicating a much higher plant-base uptake of ^{99}Tc following the post-transplanting deposition. The present results can be referred to in a radiological impact assessment for the contamination of paddy fields with ^{99}Tc by chronic or acute releases from nuclear facilities.

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