Report

The Effect of Soil Sterilization on the ^{137}Cs Transfer from Soil to Radish (*Raphanus sativus* var. *sativus*) —Transfer Experiment Involving Sterilized Soil—

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Three different types of soil were collected at each of three locations: a persimmon orchard, an ume (*Prunus mume*, so to speak, 'a Japanese apricot') orchard, and a paddy field located 50–55 km northwest from Fukushima Daiichi Nuclear Power Plant. The goal was to investigate the involvement of microbes inhabiting these soils on the behavior of ^{137}Cs . The soils were sterilized with gamma ray irradiation for 30 hours (absorbed dose of 60 kGy) or with high-pressure steam (autoclave sterilization) at 121°C for 20 minutes. A radish cultivar (*Raphanus sativus* var. *sativus*) was then cultivated in those soils for 45 days, and the harvested taproots and leaves were tested using a Ge semiconductor detector for concentration of ^{137}Cs . The result showed that the concentration of ^{137}Cs in radishes cultivated in the sterilized soils with autoclave sterilization or gamma ray irradiation were significantly higher than in those cultivated in the unsterilized soils. An increase in the plant available ^{137}Cs could be caused by NH_4^+ arisen from the multiple effects of the structural change of the soil, decomposition of organic matter, and/or extinction of the microbes by sterilization.

KEY WORDS: ^{137}Cs , sterilization, taproot uptake, radish, microbes, autoclave sterilization, gamma ray irradiation, Fukushima Nuclear Power Plant accident.

I INTRODUCTION

A large amount of radionuclides was released into the environment by the Fukushima Daiichi Nuclear Power Plant accident on March 11, 2011. Many experiments have been conducted to understand the environmental behavior of radionuclides. Of those radionuclides, ^{137}Cs is the most important radionuclide because it has a physical half-life of 30.1 years and remains in the environment for a relatively long time.

The soil-to-plant transfer coefficient (TC) is an important parameter for evaluating internal exposure to radiation. In fact a variety of TC values has been reported for radio-caesium;^{1–3)} however, the mechanism details are not well known.⁴⁾ In this report, the influence of soil sterilization on ^{137}Cs transfer from

soil to radish (*Raphanus sativus* var. *sativus*) was investigated using actual agricultural soils collected from agricultural fields located near Fukushima Daiichi Nuclear Power Plant.

II MATERIALS AND METHODS

1. Collection of soil samples

With the support of Fukushima Agricultural Technology Center, surface soil samples (0–5 cm depth) were collected at three different sites located about 50–55 km northwest from the Fukushima Daiichi Nuclear Power Plant at September 30, 2014. The first sample was persimmon orchard soil, which was a mixture of kuroboku soil with dry grass. The second was ume orchard soil, consisting of clay loam and dry grass. The third was paddy field soil, which was a very wet kuroboku soil with a slight clay fraction. The orchard soils were not air-dried, but mixed by a ceramic ball mill and passed through a 2-mm mesh sieve. The paddy field soil was air-dried and passed through a 2-mm mesh sieve. **Table 1** shows the concentrations of ^{137}Cs in those soils.

2. Sterilization methods

Autoclave sterilization and ^{60}Co gamma ray irradiation were used for sterilization of the soil samples. Those soil samples were put in each polycarbonate pot (72 × 72 × 100 mm) by 200 g wet-weight, and then sterilized by autoclave at 121°C

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Table 1 Radioactivity concentrations of ^{137}Cs in soils (Bq/kg).

Soil from >	Persimmon orchard	Ume orchard	Paddy field
Concentration	16,000 $\pm$ 15	21,000 $\pm$ 17	6,300 $\pm$ 11

for 20 minutes, or by 60 kGy gamma ray irradiation. The success of sterilization was verified by the conventional colony formation assay.

3. Cultivation experiment

The radish cultivar (*Raphanus sativus* var. *sativus*) was used in this study. Seeds of the radish (ATARIYA Co., Ltd) were sterilized using a sterilizing solution (0.05% polyoxyethylene sorbitan monolaurate and 5% sodium hypochlorite acid), and then germinated on sterilized filter paper. Germinated radish sprouts ($n=4$) were transferred to and cultivated in each aforementioned sample soil with or without sterilization for 45 days at 22.0°C and 60% humidity under long-day condition (16 h light/8 h dark, with 40 W white fluorescent light $\times$ 15 pcs). Autoclaved and deionized water was replenished to soil using a bottom watering method.

Although four radish sprouts were independently cultivated in each soil type, the harvested taproots and leaves were combined for each soil type because the taproot and leaves of an individual radish did not provide a measureable quantity of radionuclide. Each sample was subjected to a test by a Ge semiconductor detector (GEM30-70; Seiko EG&G) for ^{137}Cs .

III RESULTS AND DISCUSSION

The dry weight of radish (leaves, taproot, and their sum) and the appearance before harvesting (showing their growth activity, i. e., fresh, green, or wilted) were compiled in **Table 2**. Growth was generally improved in radishes cultured in the sterilized soils. The weight of radish cultured in the sterilized paddy field soil was also heavier than those in the non-sterilized soil.

It has been reported that sterilization methods, such as

used in this study, increases and keeps the concentration of $\text{NH}_4\text{-N}$ in soils because of extinction of nitrifying bacteria.^{5,6} Considering that radishes cultured in solutions of NH_4^+ plus NO_3^- (a ratio of $\text{NH}_4^+/\text{NO}_3^-$ is $< 10/1$, in forms of nitrogen) grew equal to or larger and heavier than radishes in solution of NO_3^- alone,⁷ the improvement in the growth observed here might be attributable to the increased concentration of $\text{NH}_4\text{-N}$ in soils.

The concentration of ^{137}Cs in plants (leaves and taproot) was increased by the sterilization, except for plants cultured in the autoclaved soil of a persimmon orchard (**Table 3**). This tendency was most noticeably observed in the ume orchard soil. For example, the concentration of ^{137}Cs in the taproot cultured in the autoclaved soil was 8-times higher than in the unsterilized soil.

As indicated in previous reports, increased concentration of NH_4 enhances the elution and availability of Cs ions adsorbed on the soil clay particles.⁸⁻¹⁰ The cessation of activity by nitrifying bacteria after autoclave sterilization would cause accumulation of $\text{NH}_4\text{-N}$ in the sterilized soil, followed by adsorption of Cs ions to particles.

It is interesting to note that the ^{137}Cs concentration tended to be higher in soils sterilized by autoclave than those subjected to gamma ray irradiation. This may be related to the findings reported by Eno and Popenoe¹¹) that autoclave sterilization is more effective for elution of $\text{NH}_4\text{-N}$ from soils than other sterilization techniques, including gamma ray irradiation and methyl bromide fumigation. Physicochemical change of the soil due its exposure to high temperature and humidity during autoclave sterilization may be responsible for this.

IV CONCLUSIONS

This study clearly demonstrated that Cs-transfer from soil to radish was enhanced by sterilization with autoclave and with gamma ray irradiation. The detailed mechanisms are not readily apparent, but the increased concentration of $\text{NH}_4\text{-N}$ made available by sterilization may be responsible for this. There is a possibility that sterilization caused a change in the

Table 2 Dry weight (g) of leaves and taproot and growth activity.

	Persimmon orchard			Ume orchard			Paddy field		
	Un-sterilized	Gamma ray	AC ⁺	Un-sterilized	Gamma ray	AC ⁺	Un-sterilized	Gamma ray	AC ⁺
Leaves	1.5	0.79	1.89	0.17	0.29	0.94	0.24	1.0	0.66
Taproot	2.3	0.74	0.36	0.25	0.12	1.0	0.13	0.51	0.45
Sum	3.8	1.5	2.3	0.42	0.41	1.9	0.37	1.5	1.1
Growth activity	+	+++	+++	+	+++	+++	+	++	++

AC: autoclaved (high pressure steam sterilization).

⁺wilted, ⁺⁺yellow, ⁺⁺⁺fresh and green.

Table 3 Concentrations of ^{137}Cs in leaves and taproot (Bq/kg).

	Persimmon orchard			Ume orchard			Paddy field		
	Un-sterilized	Gamma ray	AC*	Un-sterilized	Gamma ray	AC*	Un-sterilized	Gamma ray	AC*
Leaves	140 $\pm$ 14	410 $\pm$ 25	62 $\pm$ 15	700 $\pm$ 20	210 $\pm$ 6.7	970 $\pm$ 14	550 $\pm$ 15	2,000 $\pm$ 20	2,400 $\pm$ 27
Taproot	250 $\pm$ 10	150 $\pm$ 15	250 $\pm$ 33	390 $\pm$ 11	2,200 $\pm$ 50	3,400 $\pm$ 62	1,200 $\pm$ 31	3,200 $\pm$ 35	4,100 $\pm$ 43
Sum	210 $\pm$ 8	280 $\pm$ 10	92 $\pm$ 3	520 $\pm$ 18	790 $\pm$ 22	2,200 $\pm$ 9.6	780 $\pm$ 17	2,400 $\pm$ 18	3,100 $\pm$ 25

physical and chemical structure of the soils, but this point awaits future investigation.

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