Radioactivity in vegetation at the Fukushima area: a study on contamination by radionuclides released from TEPCO's Fukushima Daiichi nuclear power plants

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Leaves of Kudzu collected from the Fukushima Daiichi NPP accident contamination area are shown to exhibit high levels of ^{134}Cs and ^{137}Cs activity. Radioactive dusts that attached to the leaf surface were measured at significant levels (as confirmed by IP) for a sample collected in 2011, but at lower levels for samples collected in 2012. This suggests that root uptake would be considered a major source of internal contamination prior to collection of the 2012 samples. The surface and internal contamination was evaluated for two leaves of the 2011 sample and the radiocesium attached contributed to 13% and 63% of the total activity, respectively. The $^{134+137}$Cs activity of the 2012 samples showed a positive correlation with the radiation dose rate measured at the sampling points; however, the $^{134+137}$Cs activity estimated as an internal fraction still seemed high when compared to that of the 2011 sample. These results suggest that the surface absorption of radiocesium from attached dusts is likely responsible for the observed high internal activity.

Key Words: *vegetation, Kudzu, contamination, imaging plate, Ge detector*

1. Introduction

The Fukushima Daiichi nuclear power plants lost reactor cooling functions when the Tsunami struck at the east coast of Tohoku following the earthquake of March 11, 2011. This caused radionuclides from the reactors to be released into the environment and resulted in a wide deposition of radioactive contamination over Japan[1)]. The heavy deposition of radionuclides was confirmed to be in the northwest direction from the nuclear power plants in Fukushima prefecture, extending over more than 30 km within a week of the accident. At the time of the deposition, vegetative cover of the ground surface was low as it was the end of winter. The deposition of radionuclides was primarily limited to soil surface and litter covering the ground surface[2)]. However, the radionuclides deposited on the ground surface are believed to have then permeated deeper into the soil layer[3)], where they were absorbed by vegetation that emerged later in spring[4)]. Knowledge of the fate of radionuclides deposited on the ground surface is crucial for understanding the comprehensive impact of this incident on the ecosystem[5)]. To assess the radionuclides and their behavior patterns, we measured radioactive contamination in vegetation samples using an imaging plate and Ge detectors. These instruments also helped us to understand the soil-plant interaction in the ecosystem[6)]. In this paper, we present the results obtained for radionuclides by using vegetation samples collected from Fukushima prefecture.

2. Materials and methods

A sample of Kudzu (*Pueraria lobata*), was collected on June 21, 2011 from Futaba town in Fukushima prefecture (located 2.7 km from the Fukushima Daiichi NPP), while 2012 samples were collected on July 30-31 and August 1 from several points outside the 20 km zone surrounding the Fukushima Daiichi NPP. The fresh vegetation samples were transferred into plastic bags, transported to the laboratory, and refrigerated prior to analysis. Kudzu, belonging to the pea family, is a climbing, coiling, and trailing vine that is native to southern Japan and south east China. It is typically found on the slopes of mountains and its starch in root is used to make traditional Japanese cake. The control sample used for the study was collected on September 16, 2012, from Fuji town in Saga prefecture (located at more than 1000 km from Fukushima).

A photostimulable phosphor (PSP) image plate was used to record a two-dimensional image of radioactivity distribution on the fresh Kudzu samples. We used the FLA-5100 (Fujifilm Co.), which has a large 40 × 46 cm sampling area and an imaging pixel size of as low as 10 μm. Images of the 2011 Kudzu were exposed for 15 h, while those of the 2012 samples were exposed for over 60 h. In comparison, the control sample image was exposed for 120 h because of the low activity. The images were then analyzed with a computer system meant for use with the FLA-5100. The radionuclide concentrations in Kudzu were determined using Ge detectors. The sample list is summarized in Table 1 and the sampling points are shown in Fig. 1.

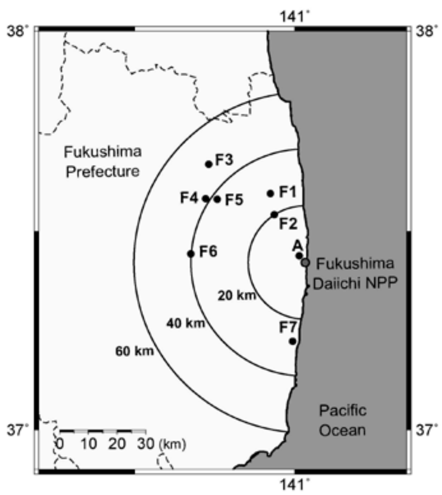

Fig. 1 Sampling points

3. Results and Discussion

(1) Radionuclide concentrations

We detected ^{134}Cs activity in all the samples collected (with the exception of the control sample), which indicated that all the sampling points within Fukushima were affected by radioactivity released from the Fukushima Daiichi NPP. The ^{134}Cs, ^{137}Cs, and ^{40}K concentrations of radionuclides observed in the samples are summarized in Table 1. The Futaba sample (collected in 2011) showed contamination with ^{110m}Ag (37±3 Bq/kg dry) and ^{129m}Te (1280±170 Bq/kg dry). However, ^{110m}Ag and ^{129m}Te were not detected in the samples collected in 2012. The 2011 sample also showed much higher concentrations of radiocesium when compared to those collected in 2012. The $^{134+137}$Cs concentrations and radiation dose rates measured with a scintillation survey meter (Aloka TCS-171) at 1 m above the ground revealed a positive correlation with regard to samples collected in 2012 (R = 0.78), as shown in Fig. 2. The $^{134+137}$Cs contamination level is clearly situated outside the correlation line for the 2012 samples. We propose that this scenario could be due to heavy concentrations of radioactive dusts attached onto the plant's surface. The IP of the Futaba sample showed many high intensity spots (Fig. 3) superimposed on the leaf, suggesting that contamination with radiocesium occurred both internally and by surface deposition. With the exception of the Akougi sample, the surface contamination measured for the 2012 samples was not clearly verified. Surface contamination data measured for the Akougi sample consisted of a small number of IP spots with weak intensity (Fig. 3).

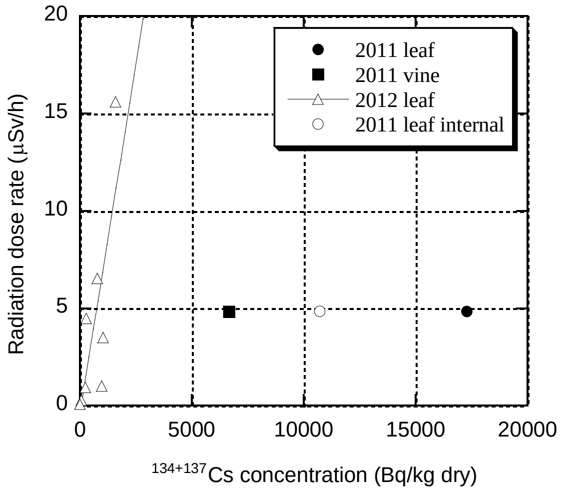

Fig. 2 Relation between radiocesium concentrations and radiation dose rates measured at 1 m above ground surface.

Table 1 Radioactivity detected on Kudzu samples.

ID	Sapling point	Sampling date	part	^{134}Cs Bq/kg dry	^{137}Cs Bq/kg dry	^{40}K Bq/kg dry
A	Futaba	June 21, 2011	leaf	8177± 15	9111±14	398 ±14
A	Futaba	June 21, 2011	vine	3136±14	3513 ±14	605 ±25
F1	Haramachiku	July 30, 2012	leaf	346 ± 15	622 ± 16	653 ± 78
F2	Odaka	July 30, 2012	leaf	393 ± 12	648 ± 12	398 ± 51
F3	Ishiusu	July 30, 2012	leaf	92 ± 9	183 ± 11	346 ± 57
F4	Mizusakai	July 31, 2012	leaf	328 ± 13	455 ± 6	803 ± 110
F5	Akougi	July 31, 2012	leaf	656 ± 17	943 ± 17	825 ± 121
F6	Tamura	July 31, 2012	leaf	24 ± 4	48 ± 3	408 ± 85
F7	Hirono	August 1, 2012	leaf	94 ± 8	141 ± 7	513 ± 72
S	Fuji	September 16, 2012	leaf	nd	nd	511 ± 55

nd: not detected * A-F7 collected in Fukushima, S in Saga.

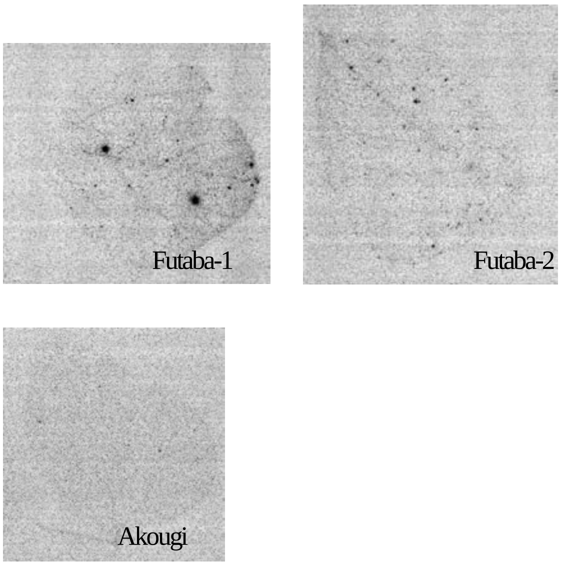

Fig. 3 The IPs of the Futaba and Akougi samples.

Kudzu loses its aerial component each fall, while the root remains alive in the winter and new buds emerge in spring. At the time of the NPP accident, no green vegetation was expected to be seen on the ground surface at the sampling points. We visited the sampling points on April 2011, and confirmed that no green vegetation was found on the ground surface. This indicated that the 2011 Futaba sample emerged at some point after April, 2011, and was not affected by the direct deposition of radioactive aerosol[7)] despite the wide range of contamination observed in the Tohoku area. While it is difficult to evaluate the relative significance of leaf surface absorption and root uptake roles for the Futaba samples, it can be concluded that root uptake was the cause of radiocesium contamination in the samples collected in 2012.

(2) Intensity in the IP of the Futaba sample

We analyzed the PSL（Photo-Stimulated Luminescence） values of the IP to evaluate radiocesium activity associated with the high intensity spots. A change of PSL values at 50 μm/pixel is depicted along a line intersecting the high intensity spot shown in Fig. 4. Our first calculations for net PSL values were made by subtracting a background PSL out of the leaf part. The net PSL values located along the line are shown in Fig. 4, where the PSL values for the leaf part are much smaller than those of the high intensity spots. To evaluate the boundary between the leaf part and spot, we obtained an average and standard deviation for the PSL values of the leaf part, and a threshold value for the boundary was taken as an average PLS value + 2 times of the standard deviation in the leaf part. We performed calculations on another line crossing the line P1-P2 at a right angle, and obtained a similar average value and standard deviation. We then treated the high-intensity spot as a circle.

We estimate that $^{134+137}Cs$ activity is internally maintained within the Futaba and Akougi samples by assuming that the PSL value is proportional to $^{134+137}Cs$ activity in/on the leaf. The PSL values for the high-intensity spots were then summed, and proportions of the PLS for the leaf part and the spots were calculated (the results are listed in Table 2). For two leaves, the sum of the PSL values for high- intensity spots is shown that they contain 13 % and 63 %, respectively, suggesting that considerable amounts of radiocesium were associated with dusts attached on the leaf surface.

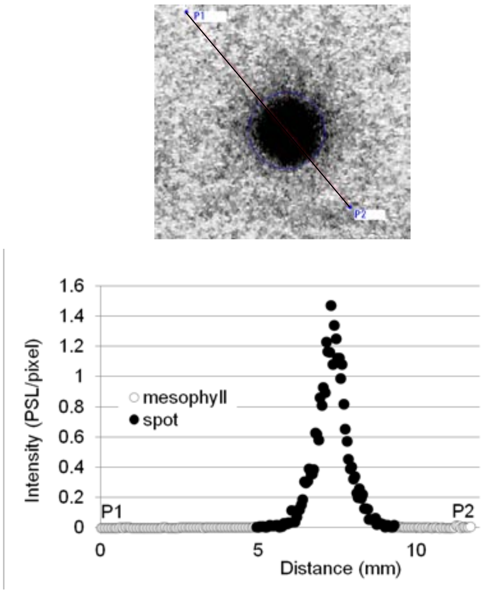

Fig. 4 The leaf IP including a high intensity spot in Futaba sample (upper) and the change in net PSL along the line P1-P2 (lower).

Among the samples collected in 2012, high-intensity spots were confirmed only on the Akougi sample. However, the sum of the PSL values for these high intensity spots also indicates that they occupied only 1 % of the surface on the Akougi sample. While surface contamination was determined to be small in this case, it was found to be more significant on the Futaba sample. The internally accumulated $^{134+137}$Cs activity for the Futaba sample, as found by averaging the two leaves, is plotted in Fig. 2. This plot also shows high radiocesium activity, and the corresponding data do not fit on the correlation line for the 2012 samples.

The vein in the leaf is clearly identified on the IP in Fig. 3, indicating higher radiocesium activity in vein than in the mesophyll. This supports the theory behind the translocation of radiocesium in plants[8], potentially as a cation, which would be well-mixed with the stable cesium. The relation between the fallout ^{137}Cs concentrations and stable Cs levels in mushrooms and plants collected in the pine forest demonstrate a positive correlation[12]. Additionally, constant ^{137}Cs/stable Cs ratio has been observed in wood xylem from pith to cambium[13, 14]. These findings suggested that ^{137}Cs is absorbed by plants, mixed with stable Cs, and then moved together. The specific activity in wood xylem was found to be uniform; however, the ^{137}Cs activity was generally high in hardwood and low in sapwood. This indicates the lateral movement of Cs through sapwood and accumulation in hardwood. The PSL values of vein and mesophyll (as estimated by IP) are given in Table 2. As opposed to mesophyll, the veins represent locations where Cs ions are believed to accumulate. However, when radiocesium is re-cycled, it returns to ground surface in the fall and is later absorbed in the spring. This soil-plant interaction is expected to continue with decreasing radiocesium activity for some period, and the decrease in activity is likely to be due to physical decay, the migration of radiocesium from the root zone, and the fixation of radiocesium to clay minerals.

Table 2 Proportion of $^{134+137}$Cs activity in Kudzu leaves

Sample	Leaf		spot
Futaba-1	37%		63%
	mesophyll	vein	
	86%	14%	
Futaba-2	87%		13%
	mesophyll	vein	
	88%	12%	
Akougi	99%		1%

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