Change of Radiocesium Concentration in Tree Leaves: Two Years' Observations Following the Fukushima Daiichi Nuclear Power Plant Accident – 14150

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ABSTRACT

Measurement of radiocesium concentrations in leaves of six tree species collected on the grounds of the National Institute of Radiological Sciences (ca. 220 km south from the Fukushima Daiichi Nuclear Power Plant) has been carried out for two years following the nuclear power plant accident to understand the concentration change with time. Three deciduous trees (Somei-yoshino cherry, Japanese chestnut, and ginkgo) and three evergreen trees (red robin, Japanese black pine, and camellia) were studied. It was found that the Cs-137 concentrations in tree leaves of each species decreased with time exponentially. The effective half-lives of radiocesium in these tree species were calculated, and the values ranged from 156 to 240 d. Although the studied trees were of different types (deciduous and evergreen), interestingly, their values were similar. As their growing climate conditions were the same, they had similar decreasing rates. The radiocesium concentrations in tree leaves and herbaceous plants were also compared for NIRS samples and samples from Fukushima Prefecture. It was found that the concentrations in tree leaves were almost the same as those in herbaceous plants for both sampling areas.

INTRODUCTION

In large areas in Eastern Japan, many trees have been contaminated with radioactive fallout from the accident that occurred at the Fukushima Daiichi Nuclear Power Plant (FDNPP) soon after the Great East Japan Earthquake on March 11, 2011. Foliar uptake and translocation of radiocesium (Cs-134: $T_{1/2}$=2.06 y, and Cs-137: $T_{1/2}$=30.17 y) have been observed in these trees. About 40-50 d after the first release of radionuclides from the FDNPP, we found newly emerged leaves of evergreen trees tended to show higher radiocesium values than deciduous trees or herbaceous plants collected at the National Institute of Radiological Sciences (NIRS) in Chiba City, Japan, about 220 km south of the FDNPP [1].

We have been carrying out the radiocesium measurements of tree leaf samples in the two years following the FDNPP accident to understand the concentration change with time. This information is important because the decreasing rates for trees in Japan might be different from those in European countries observed after the Chernobyl NPP accident due to the different tree species and climate conditions. Some reports have shown radiocesium elimination rates or in tree leaves [2-6]. Thus, in this study, we observed the radiocesium elimination trend in some Japanese tree species found at NIRS and calculated their effective half-lives.

We also compared concentrations of radiocesium in these tree leaves with those in herbaceous plants collected at NIRS and at Iitate Village in Fukushima Prefecture to see the root uptake effect in trees.

MATERIALS AND METHODS

For this study, six tree species, that is, three evergreen trees (red robin, Japanese black pine, and camellia), and three deciduous trees (Somei-yoshino cherry, Japanese chestnut, and ginkgo), were selected and leaf samples were collected and measured with a Ge detecting system (Seiko EG&G Ortec). Tree leaves had been collected from late April 2011 to August

2013, at least once a year, except for Japanese black pine. This evergreen tree species had had leaves at the time of the accident releases of radionuclides; therefore, a direct deposition effect was found for old green leaves and these results were not used in this study.

All the leaf samples were washed with tap water and then rinsed with reverse osmosis water. For April to June 2011 collections, fresh samples were measured; after washing, each sample was paper towel dried and chopped into fine pieces, and then, transferred into a container. From July 2011, each sample was oven-dried at 80˚C to decrease sample volume. Each dried sample was weighed to obtain water content, and then, broken into fine pieces. All the pieces were transferred into a plastic container for gamma-ray measurement with the Ge-detecting system using 20,000-80,000 seconds counting intervals. The water content rate was applied to the samples collected in April to June 2011 to obtain dry weight basis concentrations. Red clover samples were also collected for comparison as indicators of the bioavailable radiocesium fraction in soil.

On May 24, 2013, plant samples were collected at a sampling site in Iitate Village in Fukushima Prefecture for comparison. Samples collected were azalea (newly emerged and half a year old leaves), Japanese black pine leaves (newly emerged and one year old leaves) from a two y.o. stand and a more than 4 y.o. stand, as well as three kinds of herbaceous plants, that is, mugwort, giant butterbur and wild grass (Poaceae species, name unknown). One day after the sample collection, these samples were also treated as described above.

RESULTS AND DISCUSSION

Concentrations of both Cs-134 and Cs-137 were measured, however, due to the relatively short half-life of Cs-134 and its low concentrations in samples, only Cs-137 data are reported here. Table I shows the concentrations in leaves sampled, the sampling date and days after March 11, 2011. All leaf samples showed a decreasing trend with time. In 2011, the highest concentration was observed for camellia, 296 Bq/kg-dry. The concentrations in leaves of the selected deciduous type trees were 187-272 Bq/kg-dry, which were comparable to those for the evergreen type trees. Previously, we found that the newly emerged concentration in evergreen type trees were usually higher than other plant types [1]; however, several months later, the concentration level became similar among these tree species. Deciduous trees would absorb radiocesium deposited on the tree trunk and branch surfaces through their bark.

In 2013, concentrations of Cs-137 decreased to 8-32 Bq/kg-dry, and the concentration level was almost the same as for red clover. Since the root zones of trees are usually deeper than those of herbaceous plants, radiocesium concentration in trees is expected to become lower than that in herbaceous plants.

To compare the decreasing rate, Cs-137 concentrations in leaves need to be modeled. Figure 1 shows the concentration change in leaves of Japanese chestnut. From this result, it was clear that the concentrations in the short term following the FDNPP accident decreased exponentially. This exponential decrease could also be found in the other tree types as already reported in the previously [2-6].

TABLE I. Concentrations of Cs-137 in leaves from 2011 to 2013

Plant type	Common name	Sampling date	Days after March 11, 2011	Cs-137 Bq/kg-dry
Deciduous	Gingko	17-Nov-11	251	187±11
		12-Oct-12	581	47±6
		19-Jul-13	861	12.3±2.7
	Japanese chestnut	2-Aug-11	144	272±8
		27-Jun-12	474	81±3
		19-Jun-13	831	32±3
	Someiyoshino cherry	7-Jun-11	88	235±27
		31-May-12	447	75±5
		6-Jun-13	818	15.5±2.7
Evergreen	Red robin	15-Jun-11	96	243±27
		27-Jun-12	474	23.9±2.8
		19-Jun-13	831	15.7±3.6
	Camellia	28-Apr-11	48	296±35
		17-Feb-12	343	62±4
		22-May-13	803	30.3±2.8
	Japanese black pine	28-Sep-11	201	72±8
		6-Jun-13	818	7.9±2.7
Herb	Red clover	19-Jun-11	100	88±27
		21-Jun-12	468	35.6±3.3
		19-Apr-13	770	22.3±2.0

± shows counting error.

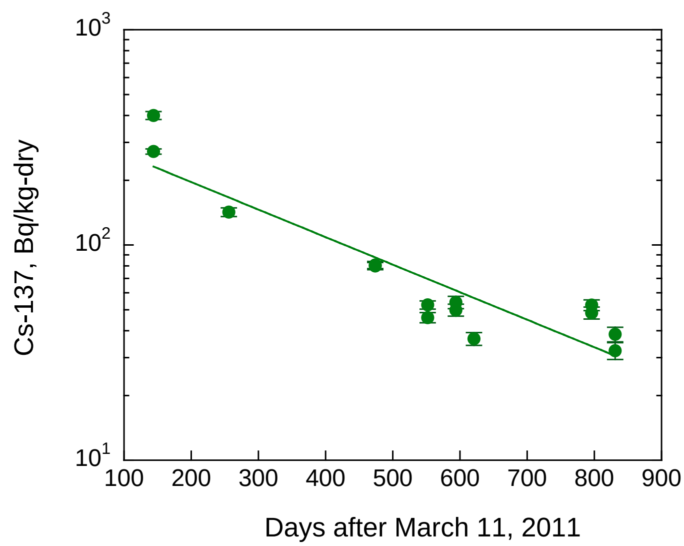

Fig. 1. Concentration of Cs-137 in leaves of Japanese chestnut plotted against time.

TABLE II. Effective half-life in tree leaves

Common name	$\mathbf{T_{eff}}$
Someiyoshino cherry	186 d
Japanese chestnut	224 d
Gingko	156 d
Red robin	185 d
Camellia	240 d
Japanese black pine	194 d

Thus, Cs-137 concentration in all tree leaves (Y Bq/kg-dry) can be modeled using the following equation;

$$Y=A \cdot \exp(-r \cdot x)$$

where, A and r are constants. Usually, a long-term effect can be explained using the sum of two exponentials, however, our sampling duration was relatively short, and we applied a single exponential model. Effective half-life (T_{eff}) of radiocesium from a tree species was then calculated using the *r* value. The calculated T_{eff} values are listed in Table II.

The T_{eff} values ranged from 156-240 d; although species were different, T_{eff} values in the NIRS collection area were similar. It was not clear why the values were similar; one thing clear was that the trees grew under the same climatic conditions (temperature, precipitation, wind and so on). For comparison, we calculated T_{eff} values for relatively short time periods, based on literature data and the values were 27 d using Swedish data [2], 125 d using Turkish data [3] and 220 d using Greek data [4]. The present data were close to that observed in Greece; however, it is necessary to clarify the reason why the T_{eff} difference was observed in different areas. It might be better to consider the climatic conditions when T_{eff} are compared among different areas.

Since concentration levels were almost the same in tree leaves and red clover sampled at NIRS, plant samples collected in Iitate Village, Fukushima on May 23, 2013 are listed in Table III for comparison. The concentrations in leaves of azalea and Japanese black pine (> four y.o. stand, one-year-old leaves) were almost the same as those observed in herbaceous plants as we observed in NIRS. However, Japanese black pine (two y.o. stand) which were grown from seed just after the accident had the highest concentration. The bioavailability change in the root zone for the last two years as well as the plant tissues that could pool radiocesium would affect the Cs concentration in the trees grown from seeds just after the FDNPP accident. These results imply that it is necessary to consider the growing conditions of the trees when their radioactivities are monitored because these conditions could cause the wide range of observation results.

TABLE III. Cs-137 concentrations in plant samples collected in Iitate Village, Fukushima Prefecture on May 24, 2013.

Type	Common name	Cs-137 Bq/kg-dry
Evergreen	Azalea, new leaves	1420±70
	Azalea, ca. 0.5 y.o. leaves	6310±200
	Japanese black pine (> 4 y.o. stand), new leaves	1270±40
	Japanese black pine (> 4 y.o. stand), 1 y.o. leaves	233±9
	Japanese black pine (2 y.o. stand), new leaves	4350±90
	Japanese black pine (2 y.o. stand), 1 y.o. leaves	21090±190
Herbaceous	Mugwort	1450±30
	Giant butterbur	723±16
	Wild grass (Poaceae species, name unknown)	7640±50

± shows counting error.

CONCLUSIONS

We observed radiocesium concentration changes in six tree species found at NIRS for the last two years following the FDNPP accident. The concentrations in leaves decreased exponentially with time. Therefore, T_{eff} was calculated for each species, and the values ranged from 156-240 d in NIRS. Although tree types were different, interestingly, the T_{eff} values were similar. This was probably due the same climate conditions affecting decreasing rates similarly for these trees. The T_{eff} values we calculated were similar to the value observed in Greece [4], rather than the values of other areas [2, 3].

When the concentration levels in tree leaves were compared with herbaceous plants, their values were close for samples collected at NIRS and in Iitate Village in 2013. Because the root zones of trees are usually deeper than those of herbaceous plants, radiocesium concentration in trees is expected to become lower than that in herbaceous plants. However, it should be noted that trees that started to grow just after the accident would pool radiocesium in their trunk and branches and thus their concentration would tend to be higher than other plants. It is necessary to consider the growing conditions of the trees used for environmental monitoring because they can possibly lead to a wide range of concentration variations.

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