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# A time dependent behavior of radiocesium from the Fukushima-fallout in litterfalls of Japanese flowering cherry trees☆

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ABSTRACT

Radiocesium ($^{134}Cs + ^{137}Cs$) concentrations, primarily derived from the Fukushima accident in March 2011, were measured in litterfalls and green leaves of Japanese flowering cherry trees (*Prunus x yedoensis* cv. Somei-Yoshino). The sampling was performed mainly during the defoliation season in 2011 and 2012 using traps to collect litterfalls before contact with the ground. The average radiocesium concentration in litterfalls in 2012 fell to one-third of that in 2011 (0.43 and 1.2 kBq kg-DW$^{-1}$, respectively). Interestingly, the concentrations in litterfalls collected in late autumn in both 2011 and 2012 (0.68 and 0.19 kBq kg-DW$^{-1}$, respectively) were significantly lower than those in litterfalls collected in the early autumn (1.7 and 1.1 kBq kg-DW$^{-1}$, respectively). In addition, the reductions in radiocesium concentrations in the litterfall were nearly synchronous with those in potassium concentrations ($p \leq 0.05$). On the contrary, radiocesium concentrations in green leaves were also correlated with potassium concentrations; however, the slopes of the regression lines between the radiocesium and potassium concentrations were very similar in the 2011 litterfalls and the 2012 litterfalls, while the slopes were significantly different between these litterfalls and the green leaves. Consequently, the correlation between potassium and radiocesium was clear but independently observable in each of the litterfalls and the green leaves. It is possible that the reduction in radiocesium concentration occurred as a part of physiological demand, a translocation of potassium from the leaves to the body/twigs.



## 1. Introduction

A large earthquake and tsunami hit northeastern Japan on 11 March 2011 and triggered a severe accident at the Fukushima Dai-ichi Nuclear Power Plant (FDNPP; located at 37.41868°N, 141.02215°E). Large areas surrounding the plant were contaminated with radionuclides consisting primarily of $^{131}I$, $^{134}Cs$, and $^{137}Cs$ (Yamamoto et al., 2012; Endo et al., 2012; Amano et al., 2012). Even in areas relatively distant from the FDNPP, such as our institute in Abiko (located at 35.87815°N, 140.02487°E; approximately 200 km SSW from the FDNPP), the contamination increased the gamma radiation dose rate to nearly 10 times pre-accident levels (data not shown). The initial radionuclide fallout was observed on 16/Mar/2011 as dry-deposition; however, the major parts of the fallout were observed on 21/Mar/2011 with rainfall in Abiko (Morino et al., 2011; Terada et al., 2012).

Woody species of plants warranted particular attention because they are effective aerosol interceptors (Bunzl and Kracke, 1988; Sokolov et al., 1990; Petroff et al., 2008; Pröhl, 2009). More than 50% of the total contamination in an evergreen forest in Fukushima was in the canopy at 5 months after the accident (Forestry Agency, 2011; Kato et al., 2012; Hashimoto et al., 2012). The removal of litterfalls from the 20 m × 20 m forest floor reduced the surrounding gamma radiation dose rate by as much as 60–70% (Tsuboyama, 2012). These findings indicate that trees and litterfalls are an important target for decontamination, especially in residential areas; however, it is unknown to what degree and how long the effect of litterfall-removal lasts. In addition, protocols for treating biological waste possibly containing radioactive materials (e.g., litterfalls and pruned twigs) need to be developed. In fact, unsorted burning of biological wastes at local incineration plants increased radio-contaminated ashes and induced a shortage of the temporal stock yards (Fukushima Prefecture, 2012). This delayed decontamination, and forced storage of raw biological waste on

*Abbreviation:* FDNPP, Fukushima Dai-ichi Nuclear Power Plant.



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private property. If radiocesium concentration dynamics in trees/litterfalls could be predicted, it would be highly useful for establishing policies for management and/or practices for gardens, green belts, orchards, and forests.

From a physiological/ecological perspective in trees, it is easily conceivable that radiocesium concentrations in trees/litterfalls fluctuate, and this may affect the redistribution of radiocesium in the environment. In fact, variations in macro and micronutrients, such as nitrogen, phosphate, potassium, and certain metals, are well documented for litterfalls (Ostman and Weaver, 1982; Duchesne et al., 2001; Hagen-Thorn et al., 2006). Retranslocation and leaching are considered to be the primary sources of variation (Duchesne et al., 2001). In addition, it is known that the phenomenon is highly element- and plant species-dependent. It is also notable that the behavior of radiocesium in plants is considerably similar to that of potassium (Ronneau et al., 1991; Zhu and Smolders, 2000; Maathuis and Sanders, 2006). However, most reports that describe radiocesium concentrations in trees demonstrate annual changes of the activity in green leaves and twigs/branches, but few describe radiocesium levels in litterfalls or refer to radiocesium recycling in plant bodies. Previous reports on litterfalls have always described the surface sediment on the forest floor but not aging leaves themselves before their dispersion to the ground (Strandberg, 1994). At the outset, we provide data showing clear, specific changes in radiocesium concentrations in litterfalls of the most popular ornamental woody species in Japanese urban areas, Japanese flowering cherry (*Prunus x yedoensis* cv. Somei-Yoshino), in 2011 and 2012. The results are discussed in relation to the physiological and ecological behaviors of radiocesium and other elements.

## 2. Materials and methods

### 2.1. The study site and target trees

The study site was in Abiko (Laboratory of Environmental Science, CRIEPI; the total area is 17.3 $km^2$), which is located approximately 200 km SSW from the FDNPP. The relative location of the study site to the FDNPP is indicated in Fig. 1A. The gamma radiation dose rate at the site was approximately 0.3–0.5 $\mu Sv\ h^{-1}$ during the sampling period. Surface soils samples of 5 cm depth around the target trees contained 0.82–3.4 kBq $kg\text{-}DW^{-1}$ of $^{134+137}Cs$ at the beginning of the sampling (Yoshihara et al., 2013). Abiko has a moderate monsoon climate (the average of 2011 and 2012 total annual rainfall and annual daily temperature are 1262.3 mm and 15.4 °C, respectively; Japan Meteorological Agency, http://www.jma.go.jp/jma/menu/report.html). The targets stood alone or as parts of colonnades and grow on bare ground. The basal soil type was pale ando soils overlaid with a thin organic layer (National Land Agency, 1983). For further descriptions related to the site, refer to the previous paper (Yoshihara et al., 2013).

The sampling targets are the most popular ornamental woody species in Japan (*Prunus x yedoensis* cv. Somei-Yoshino, Japanese flowering cherry). The distributions of the targets in the study site are plotted in Fig. 1B. The ages of the individual stands in Abiko were not specifically known; however, they were assumed to be approximately 30 y of age based on the timing of planting. The trees' heights were 9.6–14.4 m, and the diameters at breast height were 100–140 cm.

### 2.2. Sampling procedure

Litterfalls, consisting primarily of fallen leaves, were sampled in both 2011 and 2012. In detail, the sampling was performed five times each in two distinct defoliation seasons from 5 September 2011 to 31 October 2011 and from 31 August 2012 to 15 November 2012. The interval of the sampling was basically every two to three weeks. Samples from five stands were collected and independently measured with respect to the concentrations of radiocesium and eleven elements as shown below. Two box-type traps, each $1 \times 1 \times 1\ m^3$, were randomly set under each target stand to collect litterfalls before they contacted the ground. Litterfalls collected with the two traps at the same time for the same stand were mixed together and treated as one sample.

Living leaves of each target stand (i.e., green leaves) were collected on days during the vigorous growing seasons of 2011 and 2012 (6 August 2011 and 22 May 2012). The leaves harvested from at least three independent parts of each stand were mixed and treated as one green leaf sample. In addition, green leaf samples were also collected four times during the defoliation season of 2012 at the same times as the litterfall sampling procedures (from 28 September 2012 to 15 November 2012). Every litterfall and green leaf sample was dried for at least 3 d in an oven at 65 °C, weighed, and kept in the oven until further analyses were performed.

### 2.3. Radionuclide analysis

The dried samples were homogenized, weighed, and packed in screw-capped polystyrene containers (56 mm diameter × 68 mm height, U-8 container, As-one Co. Ltd., Osaka, Japan) for radiocesium concentration measurements (MEXT, 1976). We made one 3600 s measurement per sample (no replications) unless data was under the detection limit (approximately 1 Bq $kg\text{-}DW^{-1}$). For samples that were not detectable by the 3600 s measurement, another 30,000 s

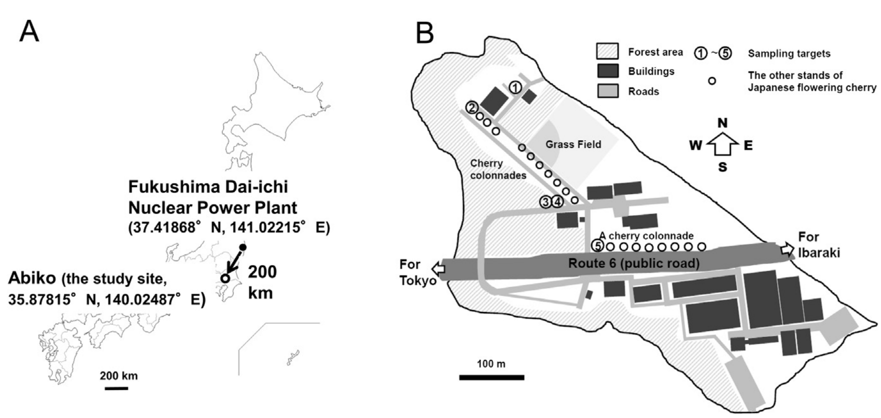


**Fig. 1.** A schematic description of the experimental site. A. The relative location of the study site from the FDNPP; B. The distributions of the target stands within the study site.