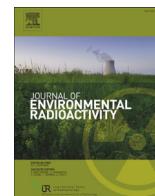




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Temporal changes in radiocesium deposition in various forest stands following the Fukushima Dai-ichi Nuclear Power Plant accident

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ABSTRACT

In this study, we investigated the transfer of canopy-intercepted radiocesium to the forest floor following the Fukushima Dai-ichi Nuclear Power Plant accident. The ¹³⁷Cs content of throughfall, stemflow, and litterfall were monitored in two coniferous stands (plantations of Japanese cedar) and a deciduous mixed broad-leaved forest stand (oak with red pine) from July 2011 to December 2012. The forest floor of cedar stands had received higher levels of additional ¹³⁷Cs deposition compared with the mixed broad-leaved stand during the sampling period. The cumulative ¹³⁷Cs deposition during the study period was 119 kBq m⁻² for the mature cedar stand, 105 kBq m⁻² for the young cedar stand, and 41.5 kBq m⁻² for the broad-leaved stand. The deposition of ¹³⁷Cs to the forest floor occurred mainly in throughfall during the first rainy season, from July to September 2011 (<200 d after the initial fallout); thereafter, the transfer of ¹³⁷Cs from the canopy to forest floor occurred mainly through litterfall. A double exponential field-loss model, which was used to simulate the removal of ¹³⁷Cs from canopies, was the best fit for the temporal changes in the canopy ¹³⁷Cs inventory.

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1. Introduction

A massive earthquake and resulting tsunami damaged the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) on March 11, 2011. The consequent accidental release of radionuclides from the damaged plant resulted in the widespread radioactive contamination of surrounding areas. In May 2011, the extent of radioactive contamination in terrestrial ecosystems within 100 km of the plant was investigated based on soil samples as part of a cooperative pilot survey involving both government and academia (MEXT, 2011a; Saito and Onda, 2015; Saito et al., 2015; Onda et al., 2015; Mikami et al., 2015). This survey demonstrated that the northwestern region of the nuclear power plant was highly contaminated by radionuclides such as radiocesium and ¹³¹I. The air dose rate and areal activity density were monitored by airborne surveys within 80 km of the nuclear power plant (MEXT, 2011b). The surveys detected widespread radioactive contamination throughout the forested areas in Fukushima and its neighboring prefectures, where approximately 70% of the land area is forested (MAFF, 2011a).

Radionuclides deposited in forested areas by either wet or dry processes encounter the canopy. Most radiocesium (>90%) deposited onto the canopy is intercepted and retained by tree leaves and branches (Hoffman et al., 1995; Pröhl and Hoffman, 1996; Kinnersley et al., 1996, 1997) and subsequently transferred to the forest floor as a result of weathering by rainwater and wind (Bunzl et al., 1989; Bonnet and Anderson, 1993; Pröhl, 2009). Some radiocesium is readily removed from plant surfaces by rainfall, but the remaining is strongly absorbed by leaf, branch, and bark surfaces (Rauret et al., 1994). However, leaf fall due to natural tree phenologies and heavy storms can accelerate radiocesium transfer from the canopy to the forest floor (e.g., Fesenko et al., 2001; Kato et al., 2012; Teramaga et al., 2014). Therefore, long-term monitoring (i.e., several years) of radiocesium transfer in throughfall, stemflow, and litterfall is essential to understand and model radiocesium behavior in forest ecosystems. However, very few studies have measured the transfer of radiocesium at the forest scale.

In a Norway spruce forest in Munich, Germany, that had been affected by radiocesium fallout from Chernobyl (total ¹³⁴Cs fallout = 20 kBq m⁻²), Bunzl et al. (1989) found that the ecological half-life of canopy-intercepted ¹³⁴Cs was 90 d for the period 0–130 d, and 230 d for the period 130–600 d following the

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beginning of radioactive fallout (the behavior of ^{134}Cs is analogous to the behavior of ^{137}Cs in a forest environment). Furthermore, Kato et al. (2012) measured the initial interception of FDNPP accident-derived radionuclides in Japanese cypress and Japanese cedar forests in Tochigi Prefecture, 150 km southwest of the FDNPP (total ^{137}Cs fallout $< 10 \text{ kBq m}^{-2}$). Canopy-intercepted radiocesium was transferred to the forest floor mainly in throughfall, but this process decreased over time; with more than half of the total radiocesium deposited remaining in the canopy 5 months after the FDNPP accident. Bunzl et al. (1989) reported that the transfer of ^{137}Cs from the canopy to forest floor decreased according to an exponential function; therefore, the retention of ^{137}Cs in the canopy can be approximated by a single exponential function in order to calculate the ecological half-life of canopy-intercepted radiocesium. In contrast to the spruce forest studied by Bunzl et al. (1989), the ecological half-life of the canopy-intercepted ^{137}Cs was found to be 620 d in the cypress forest and 890 d in the cedar forest based on approximations by the single exponential model. The longer half-life of ^{137}Cs in cypress and cedar forests indicates that the removal of radiocesium from the contaminated canopies was slower than in the spruce forest. The continuous transfer of canopy-intercepted ^{137}Cs from the canopy to forest floor causes prolonged radioactive contamination of the forest floor for many years. Therefore, long-term predictions of radiocesium transfer from the canopy to forest floor in forest environments are essential for radiocesium behavior models in forest ecosystems. Furthermore, understanding the trends of radiocesium transfer in forest environments can be useful for planning decontamination programs and radiation protection for the general population and decontamination workers, and also contribute to the interpretation and prediction of ambient dose rates in the forest environment.

The transfer of radiocesium from the canopy to forest floor during the initial phase of an accidental radioactive contamination event can be expressed using the temporal evolution of canopy inventory levels. In the past, the loss of canopy radionuclides over time (in the field) was represented by a simple exponential model assuming pulse inputs of radionuclides, such as from a reactor

accident, and their subsequent rapid removal from the canopy by hydrological processes (Milbourn and Taylor, 1965; Chamberlain, 1970; Chadwick and Chamberlain, 1970). More recently, a double exponential or offset model that considers both the rapid decrease and the subsequent slow loss has been used for demonstrating the measured radioactivity loss from vegetation canopies (e.g., Ertel et al., 1989; Kinnersley et al., 1996; Madoz-Escande et al., 2005). These models can be useful for simulating the transfer of radiocesium from canopy to forest floor during initial radioactive contamination, when the uptake of soil radiocesium by trees is minimal. However, most of these studies focused on the retention and removal of radionuclides from crops and leafy vegetables under laboratory conditions. Thus, the applicability of field-loss models to natural forest environments must be assessed by evaluating temporal changes in canopy inventory and the transfer of radiocesium to the forest floor.

In this study, we investigated the canopy interception of radiocesium by evergreen coniferous forest and mixed broad-leaved forest stands, and its subsequent transfer to the forest floor by precipitation and litterfall during the early stages of radioactive contamination following the Fukushima Dai-ichi Nuclear Power Plant accident. We also compared temporal changes in the canopy ^{137}Cs inventory with the results of previous studies using field-loss models to characterize the self-decontamination processes of the forest stands.

2. Methodology

2.1. Study site description

Our study sites are located in Kawamata Town, Yamakiya District, in the northern part of the Fukushima Prefecture (Fig. 1). From 2003 to 2009, the mean annual precipitation was 1250 mm (Yamakiya weather station, Japan Meteorological Agency (JMA)), and the mean annual temperature was 12.2 °C (Nihonmatsu weather station, JMA) in this area. Precipitation shows high seasonal variation at the study sites (Fig. 2). More than half of the

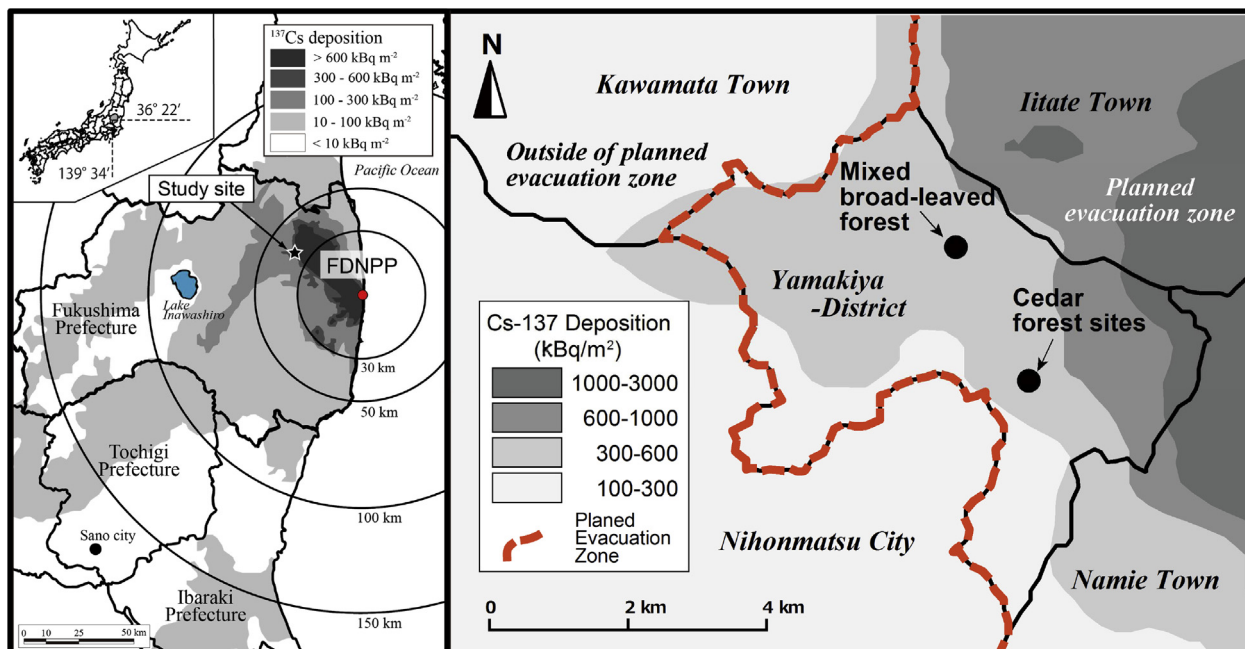


Fig. 1. Location of the study site. The ^{137}Cs deposition map is based on the Third Airborne Monitoring Survey of radioactivity by MEXT (2011b). (The values on the ^{137}Cs inventory map were corrected to the date of July 2, 2011).

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