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Short communication

# Fallout volume and litter type affect $^{137}$Cs concentration difference in litter between forest and stream environments

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ABSTRACT

It is important to understand the changes in the $^{137}$Cs concentration in litter through leaching when considering that $^{137}$Cs is transferred from basal food resources to animals in forested streams. We found that the difference of $^{137}$Cs activity concentration in litter between forest and stream was associated with both litter type and $^{137}$Cs fallout volume around Fukushima, Japan. The $^{137}$Cs activity concentrations in the litter of evergreen conifers tended to be greater than those in the litter of broad-leaved deciduous trees because of the absence of deciduous leaves during the fallout period in March 2011. Moreover, $^{137}$Cs activity concentrations in forest litter were greater with respect to the $^{137}$Cs fallout volume. The $^{137}$Cs activity concentrations in stream litter were much lower than those in forest litter when those in forest litter were higher. The $^{137}$Cs leaching patterns indicated that the differences in $^{137}$Cs activity concentration between forest and stream litter could change with changes in both fallout volume and litter type. Because litter is an important basal food resource in the food webs of both forests and streams, the $^{137}$Cs concentration gradient reflects to possible $^{137}$Cs transfer from lower to higher trophic animals. Our findings will improve our understanding of the spatial heterogeneity and variability of $^{137}$Cs concentrations in animals resident to the contaminated landscape.



## 1. Introduction

Cesium-137 fallout due to the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in March 2011 resulted in extensive contamination of the forest landscape of Fukushima and the surrounding regions (Yasunari et al., 2011; Hashimoto et al., 2012). Approximately 70% of the land area of Japan is covered by forest, and evergreen coniferous plantations and deciduous secondary forests are the most dominant forest types in Japan (Kuroda et al., 2013). In evergreen coniferous forest, a large amount of atmospherically supplied $^{137}$Cs was attached to the canopy, including foliated branches, while $^{137}$Cs fallout was deposited mostly on the forest floor in deciduous forest because of the absence of leaves in early March (Kato et al., 2012). Cesium-137 contamination deposited on forest ecosystems has further circulated within the ecosystems (e.g., Steiner et al., 2002). Hence, the movement of $^{137}$Cs may differ depending on the differences in $^{137}$Cs attachment to leaves between evergreen coniferous and broad-leaved deciduous forests (Kato et al., in press).

In forest environments, terrestrial and aquatic food webs are often structured by detrital food chains that originate from litter because of the limited light conditions under the canopies (Vannote et al., 1980; Sakai et al., 2016). Therefore, $^{137}$Cs transported with litterfall can generate bottom-up $^{137}$Cs transfers to higher trophic levels in such forest ecosystems (Sakai et al., 2016). Recent studies demonstrated that the $^{137}$Cs concentration in litter is lower in streams than in adjacent forest environments (Sakai et al., 2015). Such differences further reflect the contamination levels of animal communities in the food webs of forests and streams (Sakai et al., 2016). The findings of the previous studies indicated that $^{137}$Cs leaching from contaminated litter is the primary process influencing the dynamics of contaminants in stream ecosystems.

Cesium-137 transported with litterfall to streams can differ depending on the dominant forest types in riparian zones, because the canopies of evergreen coniferous forests were more contaminated than those of broad-leaved deciduous forests following the FDNPP accident (Kato et al., in press). Such differences in the $^{137}$Cs

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concentration between coniferous and deciduous litter may further reflect the leaching patterns in streams, because the leaching pattern may depend on the concentration gradient (Hefter and Tomkins, 2003; Sakai et al., 2016). Changes in the $^{137}$Cs concentration in litter due to leaching in streams can be high in evergreen coniferous litter because of the high initial contamination level compared with broad-leaved deciduous litter.

In addition to the litter type, the $^{137}$Cs concentrations in litter are also associated with the $^{137}$Cs fallout volume in forests. The $^{137}$Cs fallout was spatially heterogeneous from one watershed to another (Kinoshita et al., 2011). Such distribution patterns are also related to the $^{137}$Cs concentration in particulate matter in streams (Yoshimura et al., 2015). Therefore, the different $^{137}$Cs concentrations in litter due to variations in fallout volume might lead to differences in the loss of $^{137}$Cs once the litter enters stream channels in riparian zones. Thus, we hypothesized that both litter types (evergreen coniferous and broad-leaved deciduous litters) and $^{137}$Cs fallout volumes were the primary factors responsible for differences in the $^{137}$Cs concentration between forests and streams. This hypothesis was tested by investigating the $^{137}$Cs activity concentrations in litter in both forests and streams with different contamination levels.

## 2. Materials and methods

### 2.1. Study site and sampling

This study was conducted in seven stream reaches, with riparian forest affected by different levels of contamination in Fukushima (six sites) and Gunma (one site) Prefectures (Table 1, Fig. 1). The seven study sites had different amounts of $^{137}$Cs fallout, ranging from the lowest with 30–60 kBq/m$^2$ to the highest with 1000–3000 kBq/m$^2$, with all values determined by the governmental airborne monitoring in June 2012 (MEXT, 2012). Detailed methodology of the airborne monitoring can be found at: http://ramap.jmc.or.jp/map/eng/about.html. We classified the contamination levels into nine categories to enable a statistical analysis (see Section 2.3). Contamination classes 1 to 9 corresponded to the following respective fallout volumes (ranges of volume); ≤10, 10–30, 30–60, 60–100, 100–300, 300–600, 600–1000, 1000–3000, and >3000 kBq/m$^2$. Site IDs A to G corresponded to a range from high (class 8) to low (class 3) contamination (Table 1). Most of the riparian forests of our study sites were mixed forests, containing evergreen conifers and broad-leaved deciduous trees, with the exception of site B, which was covered only by an evergreen coniferous plantation. The evergreen conifer species was Japanese cedar (*Cryptomeria japonica*) in all sites, whereas the dominant deciduous tree species of the sites belonged to several families such as Fagaceae, Ulmaceae, and Sapindaceae. For sampling forest and stream litter, we selected a 50-m channel reach, with a 20 m-wide riparian zone on both sides.

Three samples of both evergreen coniferous and broad-leaved deciduous litters were collected randomly from the riparian zone and in-channel region. In the monoculture riparian forest site (site B), we only collected evergreen coniferous litter. Samples of broad-leaved deciduous litter comprised a range of families that were dominant within the sites. We selected a similar quality of litter, all with an intact shape and from the early stage of conditioning before starting physical and biological decomposition (Webster and Benfield, 1986; Sakai et al., 2016). All litter sampled from the forest floor was not attached to the mineral soil horizon. Samples in the stream were removed from submerged litter patches on the streambed. When sediment was attached to litter surfaces in streams, we gently rinsed and removed the sedimentary materials. Litter sampling from all sites (except site G) was completed in June 2014. Samples were also collected in May 2013 at sites F and G. Although duration of water soak can affect amount of $^{137}$Cs leaching from litter (Sakai et al., 2015), we assumed that the sampling criteria that control decomposition status of the litter samples aligned durations of water soak and enabled us to conduct fair comparisons of the difference in $^{137}$Cs concentrations in litter between forest and stream among the study sites.

### 2.2. Laboratory analysis

All litter samples were dried at 60 °C for 1 week and then pulverized using an electrical mill (FM-1; Osaka Chemical Co., Ltd., Osaka, Japan) for accurate $^{137}$Cs measurements (Sakai et al., 2015). Processed litter samples were packed into 100-mL plastic containers, and their dry weights and densities were measured. The activity concentration of $^{137}$Cs in the samples was determined by gamma-ray spectroscopy. Gamma-ray emissions at energies of 661.6 keV were counted using a high-purity germanium coaxial detector system coupled to a multi-channel analyzer (GCW2022 coupled to DSA1000, Canberra, Meriden, CT, USA; Ortec GEM20-70 coupled to DSPEC jr. 2.0, Ametek-AMT, Beijing, PRC). The energy and efficiency calibrations for this detector were performed using standard and blank (background) samples. For the analysis of radionuclide activity, each sample was measured for <10% of the error counts per net area counts. The geometry was held constant when counting all samples for $^{137}$Cs activity concentrations. All activities were corrected for decay according to the final sampling date (June 6, 2014) prior to statistical analysis. Our detector systems

**Table 1**
Location, site identification (ID), latitude, longitude, altitude, distance from the Fukushima Daiichi Nuclear Power Plant (FDNPP), $^{137}$Cs fallout volume, contamination class, and forest type at the study sites. The $^{137}$Cs fallout volumes at each site are taken from the governmental airborne monitoring data (MEXT, 2012), and the number of each contamination class was assigned with respect to each $^{137}$Cs deposition range shown in MEXT (2012).

| Place (Prefecture) | Site ID | Latitude (°) | Longitude (°) | Altitude (m) | Distance from the FDNPP | $^{137}$Cs fallout volume (kBq/m$^2$) | Contamination class | Forest type |
|---|---|---|---|---|---|---|---|---|
| Iitate village (Fukushima) | A | 37.63N | 140.78E | 510 | 34 km northwest | 1000–3000 | 8 | coniferous and deciduous |
| Iitate village (Fukushima) | B | 37.68N | 140.80E | 470 | 37 km northwest | 600–1000 | 7 | coniferous |
| Iitate village (Fukushima) | C | 37.64N | 140.67E | 630 | 43 km northwest | 600–1000 | 7 | coniferous and deciduous |
| Iitate village (Fukushima) | D | 37.72N | 140.79E | 370 | 44 km northwest | 300–600 | 6 | coniferous and deciduous |
| Iitate village (Fukushima) | E | 37.74N | 140.69E | 530 | 50 km northwest | 300–600 | 6 | coniferous and deciduous |
| Nihonmatsu city (Fukushima) | F | 37.60N | 140.61E | 550 | 45 km west | 100–300 | 5 | coniferous and deciduous |
| Midori city (Gunma) | G | 36.55N | 139.35E | 620 | 180 km southwest | 30–60 | 3 | coniferous and deciduous |