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Radiocesium leaching from contaminated litter in forest streams

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ABSTRACT

In Japanese forests suffering from the Fukushima Daiichi Nuclear Power Plant accident, litter fall provides a large amount of radiocesium from forests to streams. Submerged litter is processed to become a vital food resource for various stream organisms through initial leaching and subsequent decomposition. Although leaching from litter can detach radiocesium similarly to potassium, radiocesium leaching and its migration are poorly understood. We examined both radiocesium and potassium leaching to the water column and radiocesium allocation to minerals (glass beads, silica sand, and vermiculite) in the laboratory using soaked litter with and without minerals on a water column. The mineral types did not affect radiocesium leaching from litter, but soaking in water for 1, 7, and 30 days decreased the radiocesium concentration in litter by ×0.71, ×0.66, and ×0.56, respectively. Meanwhile, the 1-, 7-, and 30-day experiments decreased potassium concentration in litter by ×0.17, ×0.11, and ×0.09, respectively. Leached radiocesium remained in a dissolved form when there was no mineral phases present in the water, whereas there was sorption onto the minerals when they were present. In particular, vermiculite adsorbed radiocesium by two to three orders of magnitude more effectively than the other minerals. Because radiocesium forms (such as that dissolved or adsorbed to organic matter or minerals) can further mobilize to ecosystems, our findings will increase our understanding regarding the dynamics of radiocesium in stream ecosystems.

1. Introduction

Litter in the forested area is one of the most important basal food resources in stream ecosystems (Vannote et al., 1980). Due to shading by the forest canopy, growth of primary producers such as algae is generally limited in streams. Thus, organisms in streams primarily depend on litter as a vital energy source in forested areas (Richardson and Danehy, 2007). Once litter reaches the streams, leaching and colonization of bacterial and fungal species make litter a palatable food resource for stream macroinvertebrates (Cummins et al., 1989). Shredders, which comprise a functional feeding group of macroinvertebrates, play an important role in litter decomposition. During this decomposition process, elements in litter are assimilated by shredder macroinvertebrates and transferred to organisms in higher trophic levels such as predators. Consequently, transfer through the food web can propagate elements contained in litter components to the whole stream ecosystem.

Since the 1950s, the majority of the planet has experienced anthropogenic radiation contamination caused by nuclear bomb tests and nuclear power plant accidents (Gastberger et al., 2000; Whicker and Pinder, 2002). Radionuclides emitted from anthropogenic sources have generally been dispersed by atmospheric circulation and deposited to the land (Yasunari et al., 2011). Since 2011, radionuclide emissions due to the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident have contaminated the forest landscape of Japan. Among the emitted radionuclides from the FDNPP, radiocesium (^{134}Cs and ^{137}Cs) can induce long periods of contamination because of their long half-lives (Sakai et al., 2014). Radiocesium emitted from the FDNPP was initially deposited and absorbed to canopy layers in coniferous forests due to the presence of leaves during the major fall during early spring (Hashimoto et al., 2012; Kato et al., 2012). Litter fall from the canopy layer is the main radiocesium transport route to the ground surface (Teramage et al., 2014), along with stream channel draining within such forests.

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Contaminated substances in forested areas can be transferred to food webs in stream channels via elements absorbed in the litter. Similar to other contaminated substances such as mercury and pesticides (e.g., Driscoll et al., 2007; Walters et al., 2008), radiocesium can be transferred through the food web by processing litter and subsequent consumption by organisms in higher trophic levels. For example, Murakami et al. (2014) reported that radiocesium adsorbed to litter was consumed by detritivorous organisms both in terrestrial and freshwater ecosystems in forests. The radiocesium concentration was also observed in stream macroinvertebrates with functional feeding groups of shredders and predators (Yoshimura and Akama, 2014). These studies suggest that the order of magnitude of litter contamination is an important factor in determining radiocesium concentrations in organisms.

Contamination levels of radiocesium in litter from forest and stream ecosystems have differed significantly, even though these ecosystems were adjacent and closely linked. Murakami et al. (2014) reported that radiocesium concentrations in terrestrial litter were one to two orders of magnitude higher than in aquatic litter. Iwamoto (2014) showed that litter in stream channels had concentrations four times lower than litter on the forest floor, although similar litter was sampled in neighboring systems. Despite the different contamination levels of litter between forests and streams, possible mechanisms for radiocesium detachment remain unclear. Because cesium can behave similarly to potassium (Avery, 1996), radiocesium attached to litter potentially leaches from the litter through similar processes as potassium immediately after being submerged in water (Gessner, 1991). Therefore, we hypothesized that radiocesium in litter can be detached by water, and that their detachment level can vary based on the duration of submerged periods.

Availability of receptor substances in the water column can also be an important factor in the movement of radiocesium in aquatic environments because ionic and unstable radiocesium actively transfers to other substances available in the water column (Nagao et al., 2013). Radiocesium can be strongly absorbed to frayed edge sites of 2:1 layer silicate clay minerals (Francis and Brinkley, 1976; Nakao et al., 2008). Radiocesium fixation by clay minerals can suppress subsequent biological transfers because strongly sorbed radiocesium is not bioavailable (Rosén et al., 2006). Therefore, we hypothesized that radiocesium detachment from litter can be transferred to the other substances, and that the order of magnitude of transfer can vary depending on the type of substance.

We investigated the detachment of radiocesium as well as potassium from contaminated litter and the movement of leached radiocesium to other materials. We examined the detachment of radiocesium from litter to stream channels under the following conditions: (1) sources of water, (2) duration of submergence, and (3) presence of receptors in the water column. Radiocesium receptors can include various types of minerals. This will increase our understanding of radiocesium dynamics and can be used for future modeling in forest and stream ecosystems because radiocesium transported through litter fall and subsequent detachment in the stream can function as dominant radiocesium flows within the system.

2. Material and methods

2.1. Study site and sample collection

We collected Japanese cedar (*Cryptomeria japonica*) litter at a plantation watershed in Nihonmatsu City, Fukushima Prefecture, ~45 km from the FDNPP (37°36′N, 140°37′E). The mean air dose rate and ^{137}Cs inventory based on an airborne investigation on June 28, 2012, were 1.0–1.9 μSv/h and 100–300 kBq/m^2, respectively (MEXT, 2012). The mean annual precipitation was 1248 mm and the air temperature was 10.9 °C based on the Funehiki AMeDAS automated weather station located 18 km south of the site. The underlying geology was Early Cretaceous granite, the overstory vegetation was Japanese cedar, and the dominant understory vegetation species were *Actinidia polygama*, *Elatostema involucratum*, and *Chloranthus serratus*. Our study site was a typical Japanese headwater channel with 0.5–1 m wetted channel width consisting of sequences of steps and pools. Litter fall and its accumulation in the stream channel were common, especially during the early spring.

We collected ~2.5 kg of fresh litter from the forest floor (~30 m^2) and 25-L of stream water on February 13, 2014. The litter sample was selected based on the following methods: (1) needles of the litter were fully intact and attached to twigs, (2) the color of the litter remained light brown and had not turned dark brown or black, which would indicate decomposition, and (3) the litter was not partially buried in the soil matrix. The litter was dried at 60 °C for 1 week for obtaining quantitative mass of litter samples because water content of litter on forest floor can be variable. This initial treatment was essential for estimating experimental treatment effects on radiocesium and potassium leaching because leaching efficiency varies depending on initial water content of litter (Gessner, 1991). Although drying may reduce fungal and bacterial activities on litter, we assumed that drawing the conclusion based on the experiment can be robust. The stream water sample was stored at −20 °C until the laboratory experiments.

2.2. Laboratory experiment

We conducted an experiment that mixed litter samples with water and/or minerals to characterize the rate of radiocesium concentration decrease in litter in stream. Two types of water, distilled and stream water were used. The stream water was not filtered to assume natural stream conditions. We hypothesized that the amount of ionic and/or particulate radiocesium leached from litter may differ depending on the electrolyte concentration in the water. The pH of distilled and stream water were 6.8 and 6.7, and electrical conductivity (EC) of distilled and stream water were 0.09 and 6.15 mS/m, respectively, before the experiment. In addition to the two water types, we prepared three experimental durations, 1-, 7-, and 30-day. We hypothesized that the submerged duration may alter the order of magnitude of radiocesium leaching from litter.

We also prepared four mineral treatments with no mineral, glass beads, silica sand, and vermiculite. The glass beads, silica sand, and vermiculite were sieved to obtain a uniform grain distribution of 125–500 μm. The grain size distribution of the mineral treatment was assumed to comprise dominant particles found in pools of the study stream where litter is commonly deposited. We hypothesized that the mineral treatments may affect the quantity of radiocesium detachment from litter by mechanical scouring and the efficiency of radiocesium adsorption to minerals after detachment. Scouring between the litter surface and mineral particles could occur in samples with minerals, but not in samples without minerals. Because the abundance of micaceous minerals is one of the most important factors determining radiocesium adsorption to minerals (Sawhney, 1972; Francis and Brinkley, 1976), the adsorption efficiency may be greater in samples with vermiculite followed by silica sand, glass beads, and no minerals in relation to the mica content. The mineral treatments can result in a gradient of radiocesium movements through detachment by scouring and adsorption to mica. In these treatments, we assumed that the interaction among litter, water, and minerals occurred under natural stream conditions.