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Radiocesium immobilization to leaf litter by fungi during first-year decomposition in a deciduous forest in Fukushima

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ABSTRACT

Vast forest areas in eastern Japan have been contaminated with radio-isotopes by the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. Radiocesium (radioCs) is known to remain bioavailable in forest ecosystems for a long time, and it is necessary to terminate the cycling process to decontaminate the forest ecosystem. We observed radiocesium concentrations of leaf litter during decomposition on a forest floor where radiocesium (^{137}Cs) contamination was ~155 kBq/m^2. Litter bag experiments were conducted with newly fallen mixed deciduous leaf litter in a deciduous forest (alt. 610 m) about 50 km from the FDNPP. Litter bags were retrieved in April, June, August, October, and December 2012. Fresh litter ^{137}Cs concentration was ~3000 Bq/kg in December 2011. During the decomposition process on the forest floor, litter ^{137}Cs concentration increased rapidly and exceeded 25,000 Bq/kg after 6 months, whereas potassium (K) concentration in the litter was rather stable, indicating that radiocesium and K showed contrasting dynamics during the early decomposition phase. Nitrogen, phosphorus, and ^{137}Cs contents were positively correlated to fungal biomass, evaluated by phospholipid fatty acids in the litter during decomposition. The increase of radiocesium concentration mainly occurred during from April to October, when fungal growth peaked. Therefore, this suggests fungal translocation of nutrients from outside the litter substrate (immobilization) is the mechanism to increase radiocesium in the decomposing litter. The amount of ^{137}Cs contained in the 1-year-old decomposed leaf litter was estimated to be 4% per area of the soil-contaminated ^{137}Cs.

1. Introduction

Radioactive materials were accidentally released to the environment in eastern Japan as a result of the hydrogen explosion after mass damage to the system of the Fukushima Daiichi Nuclear Power Plant (FDNPP), following the mega-earthquake and destructive tsunami on the east coast of Japan on March 11, 2011 (Saito et al., 2014; Yasunari et al., 2011). Among the elements released, three major radioactive elements, ^{131}I ($T_{1/2}$ = 8 days), ^{134}Cs ($T_{1/2}$ = 2.07 years), and ^{137}Cs ($T_{1/2}$ = 30.17 years), were found to be widely deposited to the areas around FDNPP (Hashimoto et al., 2012). Because these contaminants will be radioactive in the environment for a long time, the most critical issue is cesium (particularly ^{137}Cs) dynamics in ecosystems and its effects on human activities. The fallout of nuclides contaminated forest canopies, then radiocesium quickly moved from the canopy to the forest floor (Kato et al., 2012a), while the mineral soil layer strongly absorbed radiocesium (Kato et al., 2012b). In Sweden, more than 90% of deposited radiocesium was found in the organic layer and surface soil in a spruce forest, 6 years after radiological contaminant fallout of the Chernobyl accident in 1986 (McGee et al., 2000). Therefore, in the forest ecosystem, most radiocesium will finally accumulate in the upper layer of soil, and will affect soil organisms over the long term (Geras'kin et al., 2008; Zaitsev et al., 2014).

The litter fall and subsequent decomposition of organic matter on the forest floor and in the soil are important processes in the

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nutrient cycling of forest ecosystems (Swift et al., 1979). These processes move substantial amounts of nutrient elements from plant to soil and these elements are used again by plants after mineralization. Soil microorganisms are primary decomposers of organic matter; soil microorganisms and the interaction between microorganisms and soil animals influences the rate and process of organic matter decomposition (Swift et al., 1979; Wardle, 2002). In the process of decomposition, concentration and mass of elements in the dead organic matter show gradual changes, and the changes are specific to each element (Salamanca et al., 1998). While carbon shows stable concentrations during the decomposition process, nitrogen and phosphorous showed increased concentrations and the actual amounts contained in the leaf litter increased compared with the amounts in the initial litter. Simultaneously, almost all potassium was lost from litter within 2 years (Salamanca et al., 1998). Because the concentration level of radiocesium contaminating forests in Fukushima is lower than the level that affects survival or reproduction of soil organisms (Garnier-Laplace et al., 2011; Zaitsev et al., 2014), biological activities in the forest floor will not change after contamination by the FDNPP accident. Trophic links between organisms might transfer radiocesium from soil to aboveground organisms (Murakami et al., 2014). Detailed study of the soil biological process in relation to radiocesium is required to understand the risk of food web contamination and human exposure to the radiocesium in the forests.

Among the soil organisms, fungi were the most effective accumulators of radiocesium when the soil was contaminated by radiocesium (Calmon et al., 2009). On the forest floor, radiocesium concentration was observed to increase during the decomposition process (Bruckmann and Wolters, 1994; Fukuyama and Takenaka, 2004; Witkamp and Barzansky, 1968). Although these studies suggested that microbial activities were related to the increase in radiocesium in the decomposing litter, there was no quantitative analysis of which microorganisms were responsible for the increase in radiocesium in the decomposing litter. In this study, we observed radiocesium and mineral element dynamics of tree leaf litter during first-year decomposition on the forest floor, and quantified bacterial and fungal biomass of the litter using the phospholipid fatty acid (PLFA) analysis.

2. Material and methods

2.1. Site description

The sampling site was in a deciduous forest in Shirainomori, Towa, Nihonmatsu, Fukushima Prefecture, Japan (37°35′N, 140°36′E, 610 m a.s.l.) (Fig. 1). The forest was naturally regenerated secondary forest, mainly consisting of deciduous oak (*Quercus serrata* Murray). A sampling plot (10 m × 10 m) was set in a section of sloping forest in December 2011. Soil type was brown forest soil (Cambisol).

2.2. Sampling and analysis

The vertical distribution of radiocesium specific activity in the soil profile was analyzed from core samples collected on 30 August 2012. The litter layer was very thin; therefore, it was removed from the ground surface prior to the soil sampling. Three replicates of soil samples were taken to a depth of 10 cm with a corer of 5.0 cm diameter (20 cm^2). All the samples were frozen at −45 °C before further processing. The vertical distribution of ^{137}Cs was examined by segmenting the cores into 1-cm layers. Soil samples were dried at 45 °C to constant weight and homogenized before transfer to plastic vials for ^{137}Cs determinations. The amount of radioactivity per area was estimated using the soil bulk density of each layer.

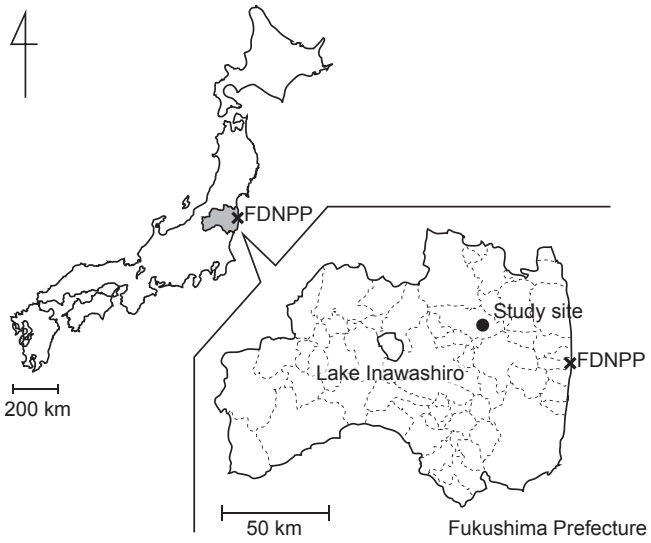

Fig. 1. Map of study site.

We used litter bags (mesh size: 2 mm and litter bag size: 25 cm × 25 cm) to monitor both mass loss and radiocesium concentration during the field experiment. Freshly fallen leaf litter was collected on 2 December 2011, and then the litter was air dried in a room after being thoroughly mixed by hand. Sixteen grams of dry weight litter was placed in each litter bag, which corresponded to 2.56 Mg/ha. Ten litter bags were set on the forest floor in 10 blocks (total 100 bags) within a 10 m × 10 m area on 17 December 2011. Litter bags from each block were retrieved in April, June, August, October, and December 2012.

At each sampling time, the litter bags were put in a plastic box and were brought back to the laboratory. Among the 10 bags, six of the litter bags were dried in an oven (45 °C for 72 h) and the other four litter bags were immediately refrigerated and stored frozen at −45 °C until the extraction of PLFA. Each dried sample (9–16 g) was ground in a Wiley mill (SKI, Retsch Muhle, Haan, Germany) and radiocesium concentration was determined by direct gamma-spectrometry using a Ge-detector for 600 s (CANBERRA GC2018). Radiocesium concentrations were shown after calculating the values at the time of field sampling considering radioactive decay. Carbon (C) and nitrogen (N) concentrations were determined by dry combustion (CN Corder, Yanaco MT-700). A 0.1 mg of dried litter sample was digested by nitric acid at 250 °C for 3–4 h, and after diluted into 50 mL using ultra pure water, the solution was filtered using membrane filter (0.45 micro m). The concentrations of calcium (Ca), cesium (Cs), magnesium (Mg), phosphorus (P), potassium (K), rubidium (Rb), and strontium (Sr) were determined using the inductively coupled plasma spectrometer (iQAP Qc, Thermo Fisher Scientific).

Microbial biomass was estimated as total extractable PLFAs. Four replicate samples of litter (0.50 g wet mass) in the litter bags were used for PLFA analysis. The litter was lyophilized and then lipids were extracted using a procedure based on those of Frostegård et al. (1991) and Niwa et al. (2008). Briefly, lipids were extracted with a one-phase chloroform-methanol-phosphate buffer, and the PLFA fraction was separated using silicic acid columns (BOND ELUT LRC-SI; Varian, Palo Alto, CA, USA) before transesterification with mild alkali and a final uptake in dichloromethane. Methyl nonadecanoate (19:0) was added to each sample as an internal standard. The fatty acid methyl esters were separated by gas chromatography with a Sherlock Microbial Identification System (MIDI, Newark, DE, USA). The fatty acid 18:2ω6,9 was used