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Radiocesium concentrations in soil and leaf after decontamination practices in a forest plantation highly polluted by the Fukushima accident☆

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ABSTRACT

Owing to the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident a vast amount of radiocesium was released polluting the land. Afterwards, a variety of decontamination practices has been done, reducing the ambient dose rates. In this study we evaluated the effectiveness of eight forest decontamination practices by means of monitoring the radiocesium (^{137}Cs) concentration in soil and leaf samples, and the daily discharge rates in ten plots during 27 months (May 2013–July 2015). A forest plantation located 16 km southwest to the FDNPP and within the exclusion area was selected. Radiocesium concentrations were analysed using a germanium gamma ray detector. The differences in radiocesium activities between the different plots were statistically significant ($p < 0.05$) and four homogeneous groups were distinguished. Tree thinning and litter removal greatly reduced the radioactivity and the two plots devoted to these practices presented the highest discharge rates of ^{137}Cs (Th + LR; 350–380 Bq/m^2 day), followed by the two Th plots (163–174 Bq/m^2 day). The clearcutting with LR and the LR plots (104 and 92 Bq/m^2 day) also had higher rates than those rates in the control plots (51 Bq/m^2 day). We only observed low rates in the two plots with matting (19–25 Bq/m^2 day). The temporal variability was explained by (i) the different rainfall depths registered during the measurement intervals (accumulated precipitation from 14 to 361 mm); and (ii) the fluctuations of the total surface coverage. The decrease trend in radiocesium concentration was high in 2013, moderate in 2014 and low in 2015 owing to the vegetation recovery after the countermeasures, thus reducing the possibility of the second pollution of the neighbouring areas. The average proportions of contribution of ^{137}Cs discharge by soil and leaf fraction were 96.6% and 3.4%.

1. Introduction

On 11 March 2011 a 9.0 earthquake and the resulting tsunami occurred in central-eastern Japan triggering, one day after, the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. After failure of the cooling systems, several hydrogen explosions damaged three of the six nuclear reactors of the power plant on March 12, 14 and 15, and affected a fourth reactor which had already been stopped (Achim et al., 2014). Published estimates suggest total release amounts of 12–36.7 PBq of ^{137}Cs (with $^{134}Cs/^{137}Cs$ approximately equal to 1.0) and 150–160 PBq of ^{131}I (Aliyu et al., 2015). Despite the bulk of radionuclides were transported offshore and out over the Pacific Ocean, significant wet and dry deposits of those radionuclides occurred on land (ca. 22%) (Morino et al., 2011). The most affected areas are in Fukushima Prefecture (Fuk-Pref, hereafter) and in a minor way in Miyagi, Tochigi, Gunma and Ibaraki Prefectures. Initial fallout contaminated cultivated soils, forests, water bodies, residential areas, asphalt and concrete surfaces, and significant pollution still remains (Saito et al., 2014; Mikami et al., 2015). After the accident, a variety of

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decontamination practices (soil removal in paddies, forests and schoolyards, litter removal, etc.) has been done, reducing the ambient dose rates (Hashimoto et al., 2012; Saegusa et al., 2015; Yang et al., 2016). This accident caused important economic damages that include, among others, the cost of decontamination practices, and the abandonment of cultivated and residential areas (Yasutaka and Naito, 2016). Additionally, there is a significant concern about the environmental and health consequences at short, medium and long-term (Aliyu et al., 2015).

Due to the strong adsorption capability of radiocesium to soil particles and vertical soil migration, seven weeks after the accident 86% of total radiocesium absorbed in the soil profile were accumulated in the upper 2.0 cm in the cultivated soils located near the FDNPP (Kato et al., 2012a). After 5 months of the initial fallout, more than 60% of the total deposited radiocesium (^{134}Cs and ^{137}Cs) in coniferous forest plantations of cypress and cedar remained in the canopy. The half lives of ^{137}Cs absorbed in the cypress and cedar canopies were calculated as 620 days and 890 days, respectively for the period of 0–160 days. The transfer of the deposited radiocesium from the canopy to the forest floor was slow compared with that of the spruce forest affected by fallout from the Chernobyl nuclear reactor accident (Kato et al., 2012b). Between 17 days and 7 months after the accident, the transfer of radiocesium from the canopy of a coniferous forest to the forest floor via throughfall and stemflow was predominant in the first weeks. However, the contributions of hydrological pathways became less important as time passed, and the litterfall route became more important (Teramage et al., 2014a). From July to September 2011 (<200 d after the initial fallout) the deposition of ^{137}Cs to the forest floor occurred mainly in throughfall during the first rainy season, thereafter (from Sep'2011 to Dec'2012), the transfer of ^{137}Cs from the canopy to forest floor occurred mainly through litterfall (Kato et al., 2017). Teramage et al. (2014a) observed that 99% of the total soil inventory of the pre- and Fukushima derived ^{137}Cs was in the upper 10 cm in a representative coniferous forest soil. On the other hand, most radiocesium in the tree rings was directly absorbed from the atmosphere via bark and leaves rather than via roots; and xylem ^{137}Cs concentrations will not be affected by root uptake if active root systems occur 10 cm below the soil (Mahara et al., 2014).

On December 2012, 22 months after the accident, Matsuda et al. (2015) found that radioactive cesium remained within 5 cm of the ground surface at most study sites (71 sites) within a 100-km radius of the FDNPP. These authors calculated a downward migration rate ranging between 1.7 and 9.6 kg m^{-2} y^{-1}. Two years after the FDNPP accident the downward migration of radiocesium to subsurface soil (5–10 cm) was only significant (26% of the total ^{137}Cs) in paddy fields near Kawamata Town (Takahashi et al., 2015). In other land uses in the Fuk-Pref, such as abandoned farmlands and tobacco fields, pastures, meadows, mixed forest, and mature and young cedar, ^{137}Cs was strongly adsorbed by soil particles and rarely migrated downward as soluble form.

In Fuk-Pref, sediment fingerprinting approaches have confirmed an effective radiocesium migration from upstream areas to coastal plains due to rainfall and spring flood events (Lepage et al., 2016). Iwagami et al. (2017a) calculated that the total annual ^{137}Cs discharge by suspended sediment, coarse organic matter, and dissolved fractions from a headwater catchment located ~35 km northwest of FDNPP was 0.02–0.3% of the total initial deposition. The total flux of radiocesium into the Pacific Ocean from the river systems and estimated at outlet stations was significant, especially during the typhoon period during the first year after the accident (Yamashiki et al., 2014). Kinouchi et al. (2015) simulated with WEP (Water and Energy transfer Process model) and calculated the long-term change in ^{137}Cs remaining in the headwater areas near the FDNPP due to the erosion and transport of contaminated sediments. These processes will reduce the amount of ^{137}Cs remaining in the catchment to 39% of the initial amount of deposition within 30 y from 2014 on, which results in the effective half-life of ^{137}Cs to be approximately 22 years.

Since the occurrence of the FDNPP accident until now the overall concentrations of ^{134}Cs ($t_{1/2} = 2.07$ years) and ^{137}Cs ($t_{1/2} = 30.17$ years) in soils, natural vegetation and crops, runoff (suspended and dissolved particles), stream water and groundwater have decreased due to the natural decay of the radionuclides as well as due to the implementation of decontamination practices. Easily applicable decontamination methods for various school facilities in Fuk-Pref included removing topsoil from schoolyards and purification of pool water (Saegusa et al., 2015). From June 2011 to July 2013 a declining trend of dissolved ^{137}Cs concentrations in stream water and groundwater (at a depth of 30 m) was observed in headwater catchments located ~35 km north west of FDNPP (Iwagami et al., 2017b). These authors also reported that the concentration of dissolved ^{137}Cs in stream water increased temporarily during the rainfall events. In cultivated soils near Kawamata Town (Fuk-Pref), the radiocesium accumulation in rice plant decreased with time (2012–2014) in both control and decontaminated fields (Yang et al., 2016). Although decontamination practices effectively reduced ^{134}Cs and ^{137}Cs concentrations (ca. 80% lower) in tadpoles in rice paddies, radiocesium concentrations in the decontaminated surface paddy soils became 3.8 times greater one year after decontamination due to the subsequent movement of radiocesium from adjacent areas (Sakai et al., 2014).

More than 70% of the Japanese archipelago is covered by forest, of which 60% is evergreen coniferous forest (Onda et al., 2010). This is also true in the highly contaminated area ($\geq$1000 kBq m^{-2}), where 66% of the area is covered by forest (Hashimoto et al., 2012). However, specific studies on decontamination practices in forest areas affected by the FDNPP accident are less common in the literature. Hashimoto et al. (2012) suggested that removing litter is an efficient method of decontamination in forest areas in Fuk-Pref, although litter is being continuously decomposed, and contaminated leaves will continue to fall on the soil surface for several years; hence, the litter should be removed promptly but continuously before more radioactive elements are transferred into the soil. Iijima et al. (2013) tested different decontamination techniques proposed by the Japan Atomic Energy Agency (JAEA) in a forest dominated by Japanese cedar trees and fir trees located 1.3 km southwest of the FDNPP. These authors found that cutting down and removing trees in the outermost area (10-m width) of the forest as well as stripping contaminated topsoil of the forest floor and pruning the trees slightly decreased the radiocesium (^{134}Cs and ^{137}Cs) dose rates. Yasutaka et al. (2013) calculated that remove fallen leaves and litter layer of 20% area of each 100 m-mesh in forest (adjacent buildings, paddy fields and other agricultural land mesh units) areas in the Evacuation Zone and Planned Evacuation Zone (restricted zone, about 1100 km^2) in Fuk-Pref had an average decontamination efficiency between 19 and 59% in air dose rates ranging between less than 1 to more than 10 μSv h^{-1}, respectively. Recently, Cresswell et al. (2016) reported that clearing trees and applying wood chips from timber reduced dose rates by 10–15% beyond that achieved by just clearing the forest litter and natural redistribution of radiocaesium in a contaminated forest near Kawamata town (Fuk-Pref).

The basic scenario of decontamination in Fuk-Pref included forested areas within 20 m of habitation areas. Implementing decontamination of all forested areas will provide some major reductions in the external radiation dose for the average inhabitant, although decontamination costs could potentially exceed JPY16 trillion (Yasutaka and Naito, 2016). This extraordinary cost and the scarcity of specific previous publications make necessary further