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Evaluating remediation of radionuclide contaminated forest near Iwaki, Japan, using radiometric methods

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A B S T R A C T

Radiometric surveys have been conducted in support of a project investigating the potential of biofuel power generation coupled with remediation of forests contaminated with radionuclides following the Fukushima Daiichi accident. Surveys conducted in 2013 and 2014 were used to determine the distribution and time dependence of radionuclides in a cedar plantation and adjacent deciduous forestry subject to downslope radionuclide migration, and a test area where litter removal was conducted. The radiocaesium results confirmed enhanced deposition levels in the evergreen areas compared with adjacent areas of deciduous forestry, implying significant differences in depositional processes during the initial interception period in 2011. Surveys were conducted both with and without a collimator on both occasions, which modified the angular response of the detector to separate radiation signals from above and below the detector. The combined data have been used to define the influence of radionuclides in the forest canopy on dose rate at 1 m, indicating that, in evergreen areas, the activity retained within the canopy even by 2013 contributed less than 5% of ground level dose rate. The time dependent changes observed allow the effect of remediation by litter removal in reducing radionuclide inventories and dose rates to be appraised relative natural redistribution processes on adjacent control areas. A 15 $\times$ 45 m area of cedar forest was remediated in September 2013. The work involved five people in a total of 160 person hours. It incurred a total dose of 40–50 μSv, and generated 2.1 t of waste comprising forest litter and understory. Average dose rates were reduced from 0.31 μSv h^{-1} to 0.22 μSv h^{-1}, with nuclide specific analyses indicating removal of 30 $\pm$ 3% of the local radiocaesium inventory. This compares with annual removal rates of 10–15% where radionuclide migration down-slope over ranges of 10–50 m could be observed within adjacent areas. Local increases were also observed in areas identified as sinks. The results confirm the utility of time-series, collimated, radiometric survey methods to account for the distribution and changes in radionuclide inventory within contaminated forests. The data on litter removal imply that significant activity transfer from canopy to soil had taken place, and provide benchmark results against which such remediation actions can be appraised.

1. Introduction

Forests are known to intercept radionuclides following atmospheric release and dispersion from nuclear sites. With activities including maintenance, commercial logging, exploitation for wild food collection and recreational activities, radioactivity in forests presents a range of radiological issues relating to external exposure, and contamination of forest products and wild foods. There are also non-radiological issues, including those associated with perceived environmental quality, cultural, ecological and social value systems. In both cases there is a need for careful assessment of the distribution of radioactive contaminants and for management systems based on an understanding of radionuclide distribution and behaviour.

The work presented here forms part of a pilot project supported by the UK Foreign and Commonwealth Office to investigate the potential of coupling forest decontamination with biomass energy production (Dutton, 2013). As part of this project investigations

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were initiated to characterise the distribution of radionuclides within a forest near Iwaki, resulting in a radiometric survey in early 2013 prior to litter removal operations in a small test area within the survey zone. Dose rate measurements were conducted in this area immediately before and after the litter clearance operations. A repeat radiometric survey was then conducted one year after the initial work to characterise the environmental change, both in the remediated area and in adjacent areas as a result of redistribution processes.

Prior to the Fukushima accident significant areas of forest have been contaminated by nuclear weapons' testing and following nuclear accidents. The processes that govern radionuclide translocation between different compartments within forest ecosystems, and removal of radionuclides from the forest, are complex and involve multiple pathways. These processes are difficult and time consuming to measure using sampling methods, and the number of studies reported in the literature is limited. Reviews of behavioural and ecological studies (Ipatyev et al., 1999; Nimis, 1996), including transfer to and within plants (IAEA, 2010; Calmon et al., 2009), and remediation options (Tikhomirov et al., 1993; Fesenko et al., 2005; Guillitte and Willdrocht, 1993; Guillitte et al., 1993, 1994; Nisbet et al., 2009) summarise knowledge of the general behaviour of radionuclides within forest ecosystems. Specific studies are nonetheless needed to assess the behaviour in new areas, such as those affected by the Fukushima accident.

Initial behaviour has been related primarily to canopy interception followed by translocation and redistribution within the living parts of trees and their associated forest litter and soil. In the first five years following the Chernobyl accident similar levels of contamination were reported in forested and adjacent pasture areas subject to wet deposition (Tikhomirov and Shcheglov, 1994; Bunzl et al., 1989), with some differences where dry deposition mechanisms are implicated. Initial interception of up to 70–80% of activity by coniferous (predominately spruce and pine) forest canopies has been reported, with substantial transfer from canopy to litter and soil observed in Ukraine, and in Nordic Countries in the first year after deposition (Tikhomirov and Shcheglov, 1994; Ipatyev et al., 1999). Longer term behaviour is expected to be determined by nutrient recycling and exchange processes between soil, litter and rooting systems, with considerable variability on local and regional scales.

In Japan, approximately 70% of the contaminated area in Fukushima Prefecture is forested (Hashimoto et al., 2013), in areas of considerably topographic relief, with high seasonal rainfall and snow-run-off. The accident occurred in early March 2011, at a time when few deciduous species were in leaf, limiting early leaf interception and immediate translocation to evergreen species. Canopy interception factors in coniferous forests (Japanese cypress, *Chamaecyparis obtuse*, and Japanese cedar, *Cryptomeria japonica*) in Japan, determined by the comparison between activity in rainwater collected in open terrain and throughfall and stemflow over the period 11th–28th March 2011, of 92% for radiocaesium have been reported (Kato et al., 2012, 2015), whereas for deciduous broadleaf forests the majority of activity has been reported to have been deposited directly onto the ground surface (Koarashi et al., 2014). These are comparable to interception factors reported for similar forests in Europe following the Chernobyl accident; studies of Norway spruce (*Picea abies*) and beech (*Fagus sylvatica*) forests at Höglwald near Munich report interception factors of 70% and 20% respectively (Bunzl et al., 1989; Schimmack et al., 1991), data from forests near Kiev report retention coefficients of 10–50% for deciduous forests and upto 100% for pine forests (Prister et al., 1994), and Melin et al. (1994) reports interception factors for spruce (*Picea abies*) and unfoliated deciduous forests in Sweden of approximately 90% and <35% respectively.

Litterfall has been reported to be a significant process in the transfer of radiocaesium from the canopy to the ground in forests in Fukushima. Teramage et al. (2014) reports that over a 200 d period, litterfall accounted for 30% of activity transferred to the ground from the canopy of a cypress (*Chamaecyparis obtuse*) forest. Over an 18 month period, Kato et al. (2015) report that litterfall accounted for 40% of activity transfer for both young and old cedar (*Cryptomeria japonica*) stands, and 64% of activity transfer for broadleaf stands. Endo et al. (2015) also reports litterfall accounting for ~50% of transfer for deciduous forests, and 69% for cedar. These studies show a significantly greater contribution from litterfall compared to the experience in Europe. Bunzl et al. (1989) reported 7% (4.6% per year) of transfer by litter fall for Norway spruce. Bonnett and Anderson (1993) reported 13–17% transfer per year by litterfall for Sitka (*Picea sitchensis*) and Norway spruce in mid Wales.

The transfer of activity from the canopy may be expressed as a double exponential decay with decay constants, which may be expressed as ecological half lives, for fast and slow components. Studies of Japanese cedar, cypress and broad leaf forests (Kato et al., 2012, 2015; Teramage et al., 2014) have reported a fast component with decay constants equivalent to a half life of 87 d, with slow components with equivalent half lives of 390 d (broad leaf), 550 d (mature cedar), and 780 d (young cedar). Bunzl et al. (1989) reported fast and slow components with effective ecological half lives of 90 d and 230 d for spruce forests. Prister et al. (1994) reported effective half lives for a fast component of 2–5 d for several different species of tree in Kiev, with slow components characterised by half lives of 25–100 d. An experimental contamination of spruce trees (Sombré et al., 1994) resulted in fast and slow components with effective half lives of 6 d and 120 d. Conversely, other studies reported no significant long term decline in activity (with effective half lives greater than 1 y) or even a slight increase in activity (Tobler et al., 1988; Raitio and Rantavaara, 1994). These are more similar to the observations in Japanese forests than the studies with slow components with decay constants equivalent to half lives of 200 d or less.

Studies of the rate of transfer from the organic soil layers to mineral soils in Japan have reported significant differences at different locations. Mahara et al. (2014) report that soil cores collected at the Fukushima Forestry Research Centre, Koriyama, in 2013 showed that more than 99% of radiocaesium activity deposited on the ground was in the litter layer and top 2.5 cm of the soil column. In contrast, Hashimoto et al. (2013) reports that for four other sites in Fukushima Prefecture the majority of the radiocaesium had migrated to the mineral soils by 2012. For most studies in European forests, transfer from the organic to mineral soil layers was slow. In Italy, Belli et al. (1994) reported less than 2% of radiocaesium in the mineral layers, in Sweden Fawaris and Johanson (1994) report <5% of activity in mineral soils in 1991, Melin et al. (1994) reports 7% of activity in mineral soils in 1990 and McGee et al. (2000) reports 77% of activity in top, mostly organic, 10 cm of soil layers in 1992. In Switzerland, however, Tobler et al. (1988) reported that only 56% of radiocaesium activity was in the litter layers by October 1986, and on sandy soils in Denmark, Strandberg (1994) reported 20% of radiocaesium in litter layers in 1991. Despite these exceptions, it appears that in general radiocaesium has been transferred to mineral soils more rapidly in Japan than in Europe. Hashimoto et al. (2013) hypothesise that this "is a result of the relatively warm climate and heavy rainfall which lead to more rapid litter decomposition and substantially thinner organic soil layers than in many European forests".

Rapid translocation of intercepted activity into cedar and red pine sapwood and heartwood has been observed (Kuroda et al., 2013). Lower activity concentrations in a range of deciduous