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

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Abstract

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

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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 15x45 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 ± 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.

Keywords Fukushima nuclear accident, collimator, radioactivity, radiocaesium, gamma ray spectrometry

Highlights

- Radiometric measurement of the distribution of radioactivity contamination in Japanese cedar forest
- Use of collimator to evaluate forest canopy contributions
- Evaluation of remediation factors following forest litter removal
- Quantification of self remediation in control area

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 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 *et.al.* 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 & 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 & 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 & 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. Bonnet & 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 & 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 & 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 broadleaf trees compared to evergreen trees have been observed, (Yoshihara *et.al.* 2013), as have direct interception by bark and translocation in deciduous fruit trees (Sato *et.al.* 2015). These differences in the rates of processes in Japan compared to Europe following the Chernobyl and Kyshtym accidents have been attributed to differences in climate, environment, timing of the Fukushima accident and potential differences in the chemical and physical forms of radioactive releases.

Potential countermeasures for forest systems range from clear felling and ploughing to access restrictions, with a corresponding range of economic and ecological effects (Tikhomirov *et.al.* 1993, Fesenko *et.al.* 2005, Guillitte & Willdrocht 1993, Guillitte *et.al*. 1993, 1994). Another approach to forest remediation is the removal of leaf litter and surface soil layers. This is labour intensive, exposes workers to radiation dose, generates significant volumes of waste, and may also have adverse effects such as loss of habitat for wildlife, reduced soil fertility, and potentially increased soil erosion. Nonetheless it has the potential of reducing dose rates in areas of high utilisation, and potentially of intercepting forest run-off.

The removal of litter and understory has been widely adopted in Japan to remediate the edges of forests, to a distance of 20 m from roads and buildings, although examinations of the effectiveness of this approach have been limited. Within the JAEA Decontamination Pilot Project, litter removal from 11 forest sites showed reductions in dose rate of 30-50% (Nakayama *et.al.* 2015), although it is not apparent that control sites were used to compare with the experimental plots.

The majority of studies of radionuclides within forests have been based on sampling the different compartments (soil, litter, wood, leaves etc) and laboratory analysis to determine activity mass concentrations. Concentration factors between media can be obtained in this way, but it is necessary to combine such observations with estimates of the mass of each compartment to determine radionuclide inventories, and to estimate the relative importance of each part of the system to external dosimetry. Elevated platforms in combination with in-situ gamma spectrometry have been used to estimate activity distributions in parts of forest canopies (Kato & Onda 2014, Yoshihara *et.al.* 2013). These studies have mostly been conducted on small experimental plots or by sampling individual trees. There is a need for extension of to larger scales to assess the extent to which small scale processes affect the mobility of radiocaesium within entire forest systems in Japan.

Radiometric survey methods are ideally suited to such larger scale studies. While regional airborne gamma spectrometry with wide line spacings can determine overall activities per unit area and close line spaced airborne work at low altitude is capable of resolving features of 50-100 m or greater (Sanderson et al 2008), ground based radiometrics has the potential for more detailed radiometric mapping in forests, bearing in mind the potential complexities of source geometries, with activity both at ground level, and in the trees and overlying canopies. In the work reported here a collimator which modifies the angular response function of a backpack detector has been used for the first time to separate signals originating from ground and canopy sources, in an attempt to account for this aspect of the source geometry. The methods used, and results of the surveys on both occasions as presented, together with a discussion of the implications of the results for forest remediation by litter removal.

2. Methods

2.1 Site Description

The site selected for this study is a community owned forest area managed by a non-government organisation, the Friends of the Forest, at Yunodake approximately 8 km south west of Iwaki, 50 km south of the FDNPP. The location is shown in Fig. 1. Operations were conducted from the Yunodake Sansoo Lodge. The site consists of deciduous broadleaf woodland and cedar (*Cryptomeria japonica*) plantations, allowing direct comparison between deciduous and coniferous forestry within a small geographic area with minimal topographic variation between the different areas. A cedar plantation between two roads to the south of the lodge, covering an area of approximately 50x300 m, was surveyed in this work, with some additional data from the deciduous woodland between this plantation and the lodge. During the fieldwork for this project, cedar tree ring samples and fresh and fallen needles were collected to investigate ^{14}C fluxes (Xu *et.al.* 2015) and radiocaesium and ^{129}I distributions between fresh needles and the litter layers (Xu *et.al.* 2016).

2.2 Instrumentation and Spectral Analysis

Measurements were conducted using Portable Gamma Spectrometry Systems developed at the Scottish Universities Environmental Research Centre (SUERC). These systems comprise a weather proof container housing a 3x3” NaI(Tl) detector with a digital spectrometer and integrated HV supply, and a GPS receiver (Cresswell *et.al.* 2013). For this work, this was carried in a backpack with a measurement time of either 5 s or 10 s for each spectrum, corresponding to averaging the signal over a distance of approximately 2-5 m. In this work the detector head is upwards, allowing the use of the collimator (see section 2.3), with 95% of the full energy radiation originating from with 10 m of the detector in open field conditions. In forests this field of view will be reduced. In total three systems were used during two periods of field work as summarised in Table 1.

As far as possible within the constraints of the terrain, a dense survey pattern of parallel lines approximately 2 m apart was maintained. Netbook or tablet computers were used to power the systems, running custom software that continuously logged spectra and associated positional information, and conducted real-time analysis using pre-determined calibration parameters. Real-time data analyses used spectral windows with a stripping algorithm to calculate activity per unit area for ^{137}Cs and ^{134}Cs, and activity per unit mass for ^{40}K, ^{214}Bi (^{238}U decay series) and ^{208}Tl (^{232}Th decay series), and a scaled count rate above 450 keV to calculate dose rate. This method, applied to airborne measurements, has been described in numerous places including IAEA (1991, 2003), Sanderson *et.al.* (1995) and Cresswell *et.al.* (2006).

The calibration parameters for the real-time analysis, taking account of the shielding effect of the operator (Buchanan *et.al.* 2016), were validated using reference sites in Scotland and Japan (Cresswell *et.al.* 2013, Sanderson *et.al.* 2013), and apply to open field geometry without, at this stage, correcting for shielding effects from trees or contributions from activity in the canopy. Preliminary Monte Carlo simulations suggest that, for the stand density and depth profile on this site, the system will underestimate radiocaesium activity per unit area by less than 20%. The relative differences across the site will be unaffected by this effect. For natural series radionuclides, the calibration assumes a uniform depth distribution. For radiocaesium, the calibration assumes a depth distribution with a mean mass depth of 0.9 g cm^{-2}, which matches calibration sites established in Fukushima in 2012 (Sanderson *et.al.* 2013) and measurements in forests elsewhere in Fukushima Prefecture of mass depths of 0.4 – 1.0 g cm^{-2} (Takahashi *et.al.* 2015).

During the January 2013 fieldwork, two areas were defined. A small stream flows from north to south through the middle of the survey area, and the area to the east of this was defined as a control area to allow comparison with natural processes, with only normal forest management conducted in this area. The remediation work was to be conducted in the western half of the cedar plantation survey area. Measurements with the collimator were only conducted within the area planned for decontamination. Decontamination would be conducted between the surveys, by removing leaf litter and cutting back understory. This work was conducted in September 2013, with a smaller area than originally intended remediated. An area of 15x45 m at the western end of the surveyed area was decontaminated by five people in 160 person hours, with 2.1 t of material removed. Dose rates were recorded using a survey meter before and after decontamination, with the average dose rate reduced from 0.31 µSv h^{-1} to 0.22 µSv h^{-1}. It is estimated that a total dose of 40-50 µSv was incurred during this remediation work.

2.3 Collimator

To assess the potential influence of activity within the forest canopy on measurements conducted on the ground a collimator was designed to provide approximately 50% attenuation of full energy radiocaesium radiation from above the detector. By comparing sequential surveys conducted with and without the collimator it was reasoned that the magnitude of contributions from the canopy could be estimated. The collimator consists of a cylindrical plastic cap with a diameter of 200 mm and height 150 mm, with a central well of diameter 125 mm and depth 100 mm. This is fitted to the top of the detector canister, enclosing the top half of the NaI(Tl) crystal.

Laboratory measurements were conducted using a point ^{137}Cs source to determine the angular response of the backpack system, both with and without the collimator in place. The measured efficiencies are shown in Fig. 2, with fitted curves of the form $\varepsilon = a + b\cos\theta + c\sin^2\theta + d\cos^2 e\theta$ (Buchanan *et.al.* 2016). A computational model using these angular responses, assuming open field conditions, gives a reduction in full energy efficiency for ^{137}Cs gamma rays (662 keV) originating above the detector of 42%, with a 22% attenuation of gamma rays from the ground surface.

Within forests, lateral attenuation of radiation from the ground by the biomass of trees reduces the proportion of radiation entering the detector at shallow angles, and confines the field of view relative to open field conditions. This reduces the effect of the collimator on gamma rays originating from the ground, relative to open field conditions, and enhances the differential sensitivity of the two measurements to canopy contributions. Increased source burial depth will have a similar effect by narrowing fields of view. Preliminary Monte Carlo simulations developed using GEANT4 (Agostinelli *et.al.* 2003, Allison *et.al.* 2006) are consistent with the experimental measurements. Simulations of a generic forest, with activity uniformly distributed in a canopy of uniform density between 2 and 5 m above the ground surface, have confirmed the reduction in the effect of the collimator for ground radiation due to the restricted field of view. For the generic geometry considered the simulation predicts a count rate of 0.53 ± 0.04 cps (Bq m^{-3})$^{-1}$ without the collimator, and 0.30 ± 0.03 cps (Bq m^{-3})$^{-1}$ with the collimator. Thus, while variations of canopy dimensions, density, activity distribution, and local topography will influence the precise partition between collimated and uncollimated surveys, the data from open field and generic forest simulations show a 42-44% reduction in canopy originating signals, and a far lower attenuation factor for the ground signal. These differences can be exploited to apportion the radiation field at operator height between canopy and ground sources.

2.4 Mapping and regridding algorithm

The dose rate (µGy h^{-1}) and ^{137}Cs and ^{134}Cs activity per unit area (kBq m^{-2}) and natural series activity per unit mass (Bq kg^{-1}) have been mapped using a modified inverse distance weighting algorithm, with the average value for each map pixel, $\bar{A}$, given by:

$$\bar{A} = \frac{\sum_i w_i A_i}{\sum_i w_i}$$

where the summation is across all measurement values A_i within a maximum range r_{max} of the map pixel. The weight assigned to each point, w_i, is given by:

$$w_i = (r_i + \Gamma)^{-p}$$

Where r_i is the distance between the measurement point and the map pixel, Γ is a constant that flattens the distribution at small values of r_i, and p is a power. For this work, a power p=2.0, Γ=1 m and maximum range r_{max}=8 m have been used, with each pixel covering an area of 0.5x0.5 m. The combination of power and flattening constant results in 95% of the weight being carried by measurements within 4 m of the pixel, approximately corresponding to the field of view of the detector. The maximum range allows two to three measurements in any direction to be included in the weighted mean value.

To allow comparisons between data collected with and without the collimator and on different occasions, a spatial regridding algorithm is employed (Sanderson *et.al* 2004, 2008). This uses the modified inverse distance weighting algorithm to determine values for dose rate, activity per unit area or activity per unit mass in each of a grid of cells. For this work, this has been done using cells of 5x5 m, using the same parameters for the interpolation and generating the mapped data.

2.5 Correction for Snow Cover

Atypically for the location of the study site, the repeat survey in February 2014 was conducted with 5-15 cm of snow cover on the ground. The attenuation of radiation through the snow thus adding to the reduction in measured dose rate and apparent activity per unit area for this survey compared to the 2013 survey. Snow cover corrections however were conducted by comparison of apparent ^{40}K activity concentrations measured in January 2013 and February 2014. Assuming that the small remediated area had not affected ^{40}K activities, and noting that the spectral interference between the minor 1365 keV radiation from ^{134}Cs and the 1460 keV ^{40}K radiation had been accounted for spectral stripping, the snow depth was estimated from the ratio of ^{40}K activity concentration measured in 2013 and 2014, as follows:

$$A_{2014} = A_{2013} e^{-\mu d}$$

where *d* is the mass depth of snow and μ the mass attenuation coefficient of water at 1460 keV. The mass attenuation coefficient for water was determined from elemental mass attenuation coefficients (Storm & Israel 1970) as 0.00574 m^2 kg^{-1}, consistent with the value calculated from the mass attenuation coefficient for water at 1500 keV given by NIST as 0.00575 m^2 kg^{-1} (Hubbell & Seltzer 2004).

The mass depth of snow was determined through the regridded data sets, and then used to determine snow-corrected activity per unit area for the later survey for ^{137}Cs and ^{134}Cs respectively using the mass attenuation coefficients for water at 662 and 795 keV, and dose rates assuming the contribution from natural sources is insignificant. It is recognised that other erosional or accumulative landcover changes between the two surveys have the potential to compound snow cover effects, although their magnitude is expected to be small in most parts of the area.

3. Results

3.1 January 2013 Results

Figure 3 shows the results of the January 2013 surveys, with maps of the activity per unit area for ^{137}Cs and the gamma dose rate. Caesium-134 activity per unit area shows the same distribution as ^{137}Cs, with an activity ratio of 0.68, and the corresponding maps are not shown. These maps show relatively low levels of ^{137}Cs activity per unit area and dose rate on the road (0.10-0.25 µGy h^{-1}, 20-40 kBq m^{-2}) and the deciduous forestry to the north of the road (0.10-0.20 µGy h^{-1}, 15-40 kBq m^{-2}) compared to the cedar forestry south of the road (0.20-0.40 µGy h^{-1}, 40-90 kBq m^{-2}). Over most of the cedar forestry, the deposited activity concentration is relatively uniform (60 ± 10 kBq m^{-2}). Lower levels of deposited activity are observed at the western edge of the forest (30-40 kBq m^{-2}) and near a stream in the middle of the area marking the edge of the control area (40-50 kBq m^{-2}). An area of higher deposited activity (70-90 kBq m^{-2}) is observed in the control area, on a slightly elevated area of ground.

Comparison between data collected with and without the collimator (Fig. 3 and Table 2) shows very small, less than 5%, reductions in estimated dose rate and radiocaesium activity per unit area using the collimator.

3.2 February 2014 Results

Figure 4 shows the snow depth calculated for each 5x5 m cell common to both the 2013 and 2014 surveys, calculated from the difference in ^{40}K count rates. Generally, snow depth in the cedar forest ranged from 20-120 kg m^{-2} (5-30 cm), with the greater depths generally on the more level ground and in hollows, and shallower depths on the more steeply sloping sections of the area. The level, open ground near the Yunodake lodge, outside the study area, had the deepest snow cover (120-180 kg m^{-2}). The uncertainties on the snow depth for individual 5x5 m cells are typically 10-20%. This is the dominant source of uncertainty in the correction of the measured radiocaesium activity per unit area to account for snow attenuation.

Figure 5 shows the ^{137}Cs activity per unit area for the 2014 survey after accounting for snow attenuation. A dose rate is calculated using conversion factors for natural and anthropogenic activity after snow correction, and is also shown in Fig. 5. The pattern of the activity distribution is very similar to the 2013 maps (Fig. 3), with a reduction evident at the western end of the cedar forestry where litter and soil removal had taken place.

Data collected with the collimator in (Table 2) shows slightly smaller, 5-15%, estimates of dose rate and radiocaesium activity per unit area compared to data collected without.

3.3 2013 vs 2014 Comparisons

Figure 6 shows the ratio of ^{137}Cs activity per unit area and dose rate between the two surveys, accounting for physical decay and snow attenuation. For much of the cedar forest surveyed, these ratios lie in the range of 0.7-0.9. The lower parts of the slopes along the southern edge of the forest and the small stream valley in the middle of the survey area show increased activity per unit area and dose rate, implying downslope migration of activity within the forest. Together these indicate that activity has migrated within the forest system over distances of 10-50 m from higher to lower elevation. The processes resulting in this downslope migration would have been ongoing since the initial deposition, and therefore it would be expected that in the 2013 data (Fig. 3) a slight elevation in ^{137}Cs activity per unit area would already be apparent. However, the increases measured here of 5-10% correspond to 2-5 kBq m^{-2} ^{137}Cs which is less than the range of each colour in Fig. 3, and much less than the variation in initial deposition measured in this work. The area to the western end of the forest which had been remediated shows significantly larger reductions in ^{137}Cs activity per unit area and dose rate, with ratios in the range 0.5-0.7.

Comparison between the mean activity per unit area and dose rate between the remediated area and the control area (Table 3) show reductions in radiocaesium of 10-15% in the control area and 30% in the remediated area. Reductions in dose rates (which also includes natural radioactivity) are slightly smaller, with remediation reducing dose rates by 24% compared to 11% reductions in the control area due to Cs migration.

4. Discussion and Conclusions

The distribution and evolution of radiocaesium and dose rate in a cedar plantation and adjacent deciduous forest near Iwaki, Japan, has been evaluated on two occasions, before and after a trial remediation experiment.

The first survey in January 2013 has shown the variability of radiocaesium deposition within this area, in particular the marked difference between the deciduous and evergreen areas, with the activity per unit area measured in the cedar plantation 2-3 times greater than that measured in the adjacent deciduous woodland in a similar topographic setting. This suggests that for this site the combination of interception by the canopy and direct deposition onto the ground was significantly greater for the cedar plantation compared to the deciduous woodland. Earlier studies comparing deciduous and evergreen forestry have shown varying results. Some of these studies have used forestry in different locations and topographic contexts, and others have reported data in activity concentrations (Bq kg^{-1}) which requires a full mass balance if inventories are to be calculated. Studies of forests following the Chernobyl accident have shown that for wet deposition there was no significant difference in deposited activity per unit area in forests compared to surrounding areas, whereas significantly elevated deposition attributed to dry processes has been reported for forested areas (Tikhomirov & Shcheglov 1994). Studies of individual trees standing alone or at the margins of small forests at Abiko, Chiba Prefecture conducted in August 2011 showed enhanced deposition in the foliage and soils below evergreen trees (cedar, pine and cypress) compared to deciduous (cherry, chestnut, sycamore and maple), expressed as Bq kg^{-1} dry weight, in a location with initial deposition by dry processes, but with the majority of deposition associated with rainfall (Yoshihara *et.al.* 2013). These observations were attributed to the timing of foliar expansion, with increased interception by the developed evergreen needles followed by transfer of intercepted activity to the ground by weathering. Similar differences in activity per unit mass for foliage in deciduous (mixed broadleaf woodland with some evergreen species) and evergreen (cedar) trees have been observed between July 2011 and February 2012 at Yamakiya, 40 km north west of FDNPP, although uncalibrated ^{137}Cs count rates at ground level do not show any pronounced differences between deciduous and evergreen trees (Kato & Onda 2014).

The studies at Yamakiya were conducted in forest of mixed beech and pine with a stand density of 2500 ha^{-1}, and young and mature cedar stands with densities of 3300 ha^{-1} and 1200 ha^{-1} respectively. Studies of orchards have also shown increased interception by evergreen trees compared to deciduous species (Sanderson et.al. 2013). In this study, the difference between deciduous and evergreen areas is similar to that observed by Yoshihara et.al. (2013), but inconsistent with the ^{137}Cs count rate data of Kato & Onda (2014). The deciduous forest at Yunodake has a lower stand density (approximately 1000 ha^{-1}) compared to Yamakiya, and does not include evergreen species. The data from this study suggests interception behaviour in the deciduous areas similar to the stand alone trees and forest edges at Abiko than the denser mixed forestry at Yamakiya. These observations are consistent with predominantly dry depositional processes being functions of stand density as well as tree species, with higher stand densities resulting in increased turbulence and reduced average airflow rates enhancing interception by trees and direct deposition to the ground surface.

Data collected using the collimator indicates that activity in the canopy of this forest produces a very small signal in the detector compared to activity on the ground. This could be a combination of significantly greater activity on the ground compared to in the canopy, the greater source to detector separation for activity in the canopy and attenuation of radiation by the canopy. Based on preliminary Monte Carlo simulations, the observed reduction in count rate with the collimator is consistent with 1-10 kBq m^{-3} ^{137}Cs in the canopy, depending on the canopy density and the activity distribution, accounting for 10-40% of the total inventory in the forest. Thus, source to detector distance and canopy self-attenuation are the dominant factors in reducing the influence of activity in the canopy on measurements conducted at ground level and the dose rate. Measurements of fresh fronds also show that these contained radiocaesium and ^{129}I, confirming that significant activity was retained in the canopy in 2013 (Xu *et.al.* 2016). Post-Chernobyl studies reported that activity intercepted by pine and birch forest canopies was transferred rapidly to the litter and soil layers, with more than 90% of the inventory in these layers after one year (Tikhomirov & Shcheglov 1994, Ipatyev *et.al.* 1999). Thus, a slower transfer from the canopy to the litter and soil than observed in the post-Chernobyl studies is implied by this data. This is consistent with other studies of evergreen forestry in Japan which has also shown longer residence times for activity in the canopies than was observed following Chernobyl (Kato *et.al.* 2012, 2015). Although radionuclides retained in the canopy do not significantly contribute to doses received by people using the forest, there may still be significant activity in the canopy that will eventually transfer to the ground, through shed needles and weathering, where it will contribute more significantly to dose rates.

An area of 15x45 m was remediated by members of the Iwaki Friends of the Forest community group, with leaf litter and understory removed by hand. The work took 160 person hours, generating 2.1 t of waste and incurring a total dose of 40-50 μSv. Measurements with a survey meter immediately before and after decontamination showed a reduction of 29% (0.31 to 0.22 μSv h^{-1}). The survey results show a reduction in radiocaesium in the remediated area of 30 ± 3%, with a reduction in dose rate of 24 ± 2%, after accounting for the physical decay of ^{134}Cs. Litter removal thus shows a beneficial, though in this instance moderate, effect on dose rate. After removal of the litter and understory, 70% of the radiocaesium remains in the environment.

Measurements with the collimator indicate that activity in the canopy has a very small impact on measurements at ground level, and therefore the majority of this measured activity is in the surface soil, having already migrated into the soil by the time this decontamination experiment had been conducted. Activity migrates from the canopy to the litter and soil via several processes. Activity in the canopy is washed out by rain, through a combination of throughfall and stemflow, with the majority of the activity transferring to the litter or soil. The shedding of leaves and needles adds contaminated material to the litter. Studies of other cedar forests have shown that throughfall dominates over litterfall, with stemflow being a minor contribution, with overall loss of radiocaesium in the canopy of mature cedar characterised by a double exponential with a rapid component with an 87 d half life and a slower component with a 550 d half life (Kato *et.al.* 2015, Loffredo *et.al.* 2014, Teramage *et.al.* 2014). Decomposition of the litter results in a transfer of activity to mineral soils. The measurements reported here are consistent with observations following the Chernobyl accident, where it was noted that >70% of the deposited activity had migrated from the litter layer to mineral soils within 2 years (Tikhomirov & Shcheglov 1994, Ipatyev *et.al.* 1999). Studies at Otama, 60 km west of FDNPP, have shown a more rapid migration from the litter to mineral soils, which it is hypothesised is a result of the relatively warm climate and heavy rainfall resulting in more rapid litter decomposition (Hashimoto *et.al.* 2012, 2013).

As the time since the accident increases, the reductions in radiocaesium inventories that can be achieved by the removal of forest litter will decline as radionuclides continue to migrate into the mineral soil layers. Hashimoto (2012) notes that 30-40% of the litter in Japanese forests will be decomposed each year, with the rate increasingly exponentially with temperature. The observations here, with approximately 30% of the activity retained in the forest litter after two summers, are consistent with this decomposition rate. Further reductions in radiocaesium inventory would require removal of soil, needing additional labour with associated dose to the work force, generating larger quantities of waste, and potentially resulting in increased ecological degradation. The effectiveness of litter removal in reducing dose rate is thus greatest when applied as soon as possible after deposition, it has been suggested (Tikhomirov et.al. 1993, Hashimoto 2012) that litter removal is credible remediation method for the first 2-3 years after deposition. Since a significant proportion of the inventory is retained in the canopy, accumulation of litter after remediation will increase concentrations on the ground. Subsequent removal of this litter may also result in a small additional dose rate reduction. It was observed in European forests that radiocaesium activity concentrations in fast growing, shallow rooted understory were greater than in the trees (Ipatyev *et.al.* 1999). Thus, if suitable plants can be identified that will grow well in Japanese forests without additional ecological problems, phytoremediation using such plants with regular clearing of the above ground plants would also result in small reductions in the radionuclide inventories of forests.

The control areas showed very significant redistribution of activities within the 12 month period under study. Natural extraction rates for many of the forest areas, particular on higher slope angle areas, were significant and lead to self remediation in certain places. Other areas with low slope angles have retained greater proportions of the activity, and there are identifiable sinks within the study area. The magnitude and distances of the redistributions implied by this study are significantly larger than would be suggested from other studies. Field monitoring of 5x22 m plots has shown 0.1% of the radiocaesium per year extracted by soil erosion (Yoshimura *et.al.* 2015). Studies of 3 m^2 plots in four different forests showed a maximum of 1.1 ± 0.5% ^{137}Cs wash off over 6 months in cypress forests, characterised by little understory, but with cedar forests showing 0.1 ± 0.1% ^{137}Cs extraction (Nishikiori *et.al.* 2015). As previously noted, an over estimation of the attenuation due to snow cover in the 2014 survey would result in an apparently larger loss of ^{137}Cs.

It is clearly important to compare remediated and non remediated areas with each other, in addition to performing time series analysis of repeat surveys if the specific impact of remediation is to be reliably established under dynamic environmental conditions. While there are studies of remediation factors from both adjacent areas (eg: the Fukushima University campus, Sanderson et.al. 2013) and time series analysis (eg: the JAEA Decontamination Pilot Project, Nakayama *et.al.* 2015), it is recommend that both approaches be combined, and that authorities who specify and evaluate remediation in future radioactive contamination take consideration of the importance of control areas.

The current situation in Japan, where approximately 70% of the contaminated area is forested (Hashimoto *et.al.* 2013), has created difficult choices for communities in balancing decisions about future management against radiological, ecological and social considerations. Remediation of more than a small fraction of this area is logistically impractical, and so any remediation activities will need to be targeted to priority areas where the maximum benefit can be gained. Radiometric methods may be useful in identifying these areas and evaluating the effectiveness of remediation. Given the long half lives of the remaining contaminants there is a need for longer term studies in order to improve knowledge and understanding of behaviour on decadal timescales. There may be opportunities for utilising the 30 year old deposition from Chernobyl in European and UK settings to learn more about these long term rates.

This study was conducted as a contribution to a project on biomass harvesting, coupling low carbon energy production with phytoremediation (Dutton 2013). It is well known that wood ash concentrates alkali and alkaline earth elements. This was known by medieval glass makers in Europe who used wood ash in making high refractive index glass (Geilmann et.al. 1955, Turner 1956, Sanderson et.al. 1984), analysis of wood ash produced from trees in the vicinity of glass making sites to determine the ranges of alkali and alkaline earth metal concentrations (Sanderson & Hunter 1981) has shown that potassium concentrations of approximately 10% in wood ash are typical, corresponding to ^{40}K concentrations of approximately 3 kBq kg^{-1}. Therefore the use of contaminated forest materials for such purposes may present management issues for the ash generated. A Swedish study, 10-20 years after the Chernobyl accident, on biofuel contamination (Hubbard & Möre 1998) concluded that ash contaminated with 5 kBq kg^{-1} ^{137}Cs returned to the land would result in annual doses of 0.1-0.5 mSv to people occupying that land. This led to radiation safety regulations on the management of contaminated ash (SSM 2012), which stream ash according to activity concentrations. Ash with concentrations of ^{137}Cs below 0.5 kBq kg^{-1} may be recycled onto forestry or arable land, ash above 10 kBq kg^{-1} must be safely disposed of, ash with intermediate activity concentrations may be used for construction or landscaping provided there is a minimum of 20 cm covering, the dose rate is less than 0.5 μSv h^{-1} and there is protection against leaching. The Swedish Radiation Safety Authority (SSM) have produced a map of areas where concentrations in ash may exceed 10 kBq kg^{-1}, areas of initial ^{137}Cs deposition in excess of 50 kBq m^{-2}, using national airborne survey data sets (Karlsson pers.comm). While some work will be needed to assess the applicability of these studies to Japanese contexts, it is noted that the Iwaki study site would be within the area where ash from biofuel utilisation may exceed 10 kBq kg^{-1}. Airborne monitoring has shown that other areas of Fukushima Prefecture have significantly higher deposition (MEXT 2011), with recent detailed airborne measurements of forested areas to the north west and south west of the FDNPP site (Sanderson et.al. 2015) identifying areas with average ^{137}Cs activity per unit area of approximately 400 kBq m^{-2}, with areas in excess of 600 kBq m^{-2}. If similar relationships to those in Sweden apply, ash from any biofuel utilisation of these areas is likely to require repository disposal.

Despite the environmental challenges of ash management, there are positive aspects to the idea of using biomass energy and selective clearance and replanting as a remediation strategy for contaminated forests. If the canopy/litter/soil exchange of the initial deposition is retarded, as suggested by this work, there may be a favourable time window before root uptake establishes further pathways for contamination of actively growing wood. Compared to the 10-20 years since deposition of the Swedish studies, forest materials harvested in Japan in the next few years may produce significantly lower activity concentrations in ash. The ecosystem dynamics are potentially complex and further work will be needed to more fully assess this potential, including detailed studies of the distribution of activity between the canopy, litter and soil. In addition to micro-scale studies of samples in laboratories, regional scale airborne radiometric methods and detailed ground-based collimated radiometrics have roles to play in further understanding the dynamics of radionuclide contamination in these important ecosystems.

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Figure 1: Location of the Yonadake study site, showing the Sansoo Lodge and the area of cedar forestry surveyed. Aerial photograph © 2015 Google. Image © 2015 DigitalGlobe.

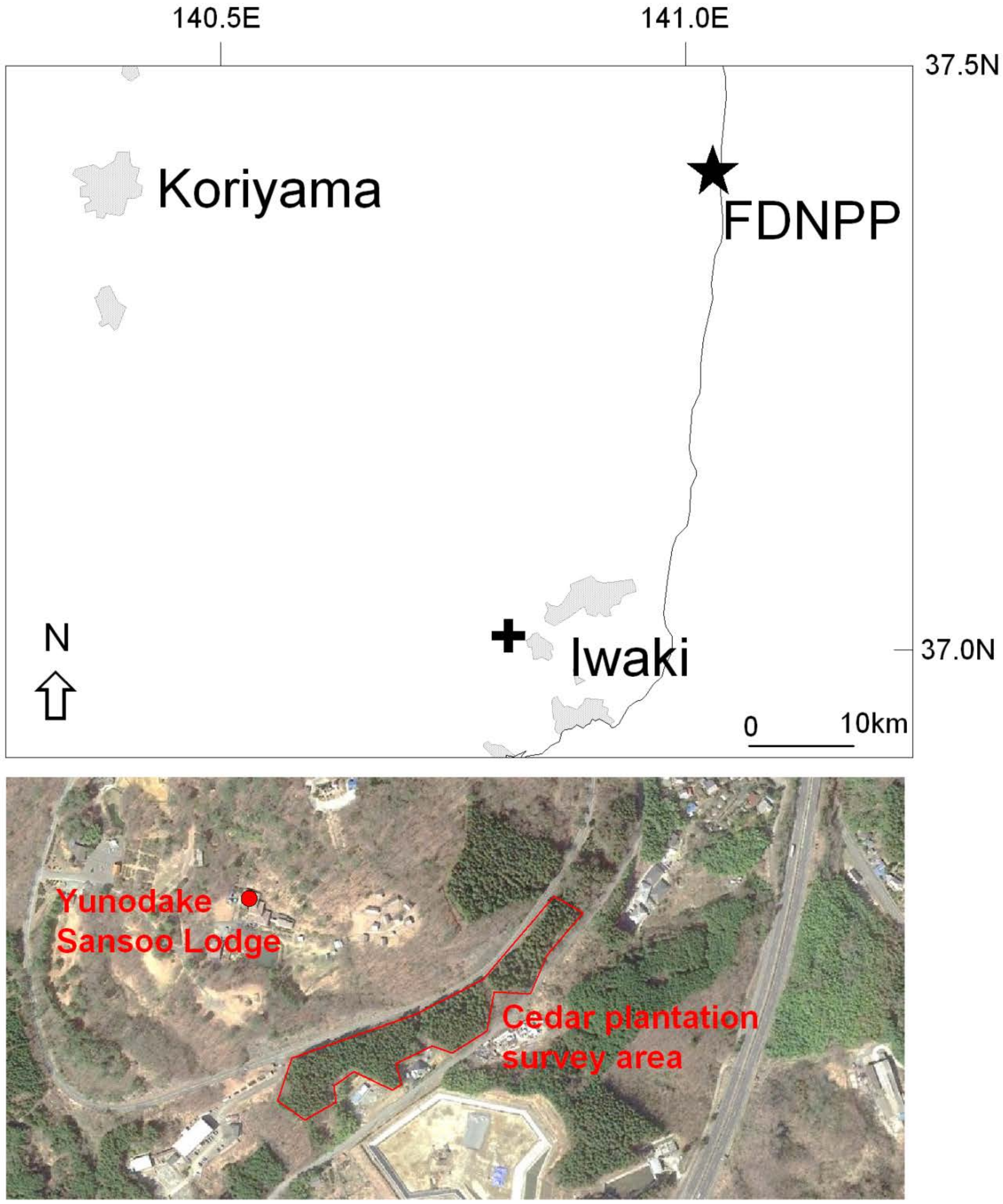


Figure 2: Angular response of the backpack system measured in the laboratory using point sources, for the standard system (red) and with the collimator (blue).

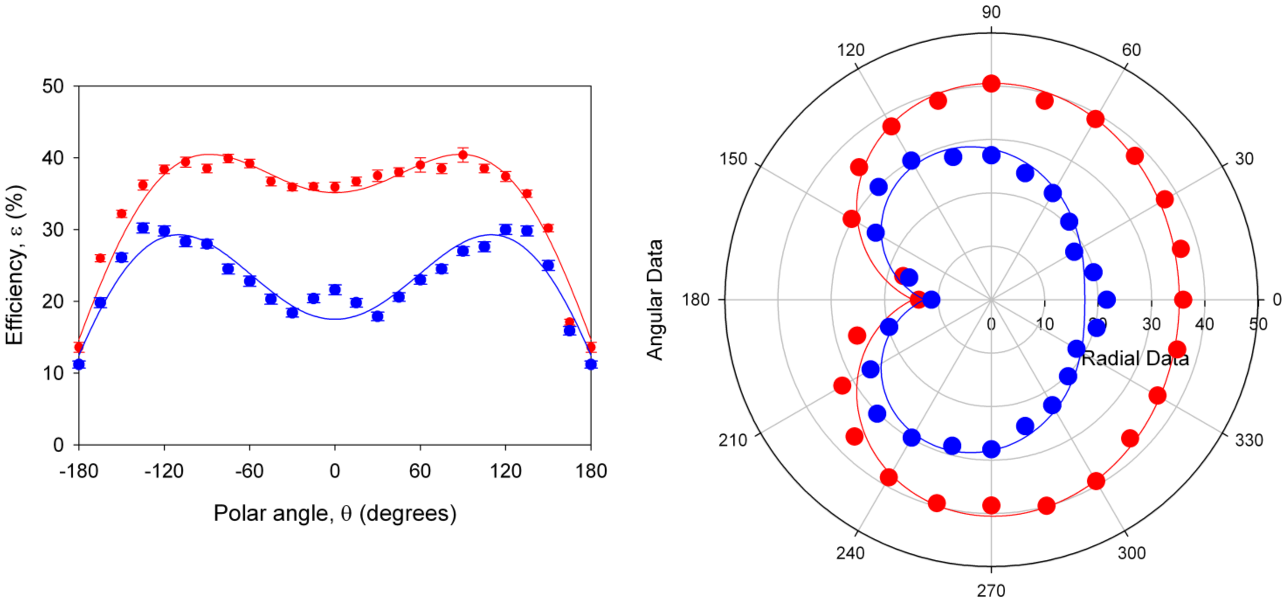
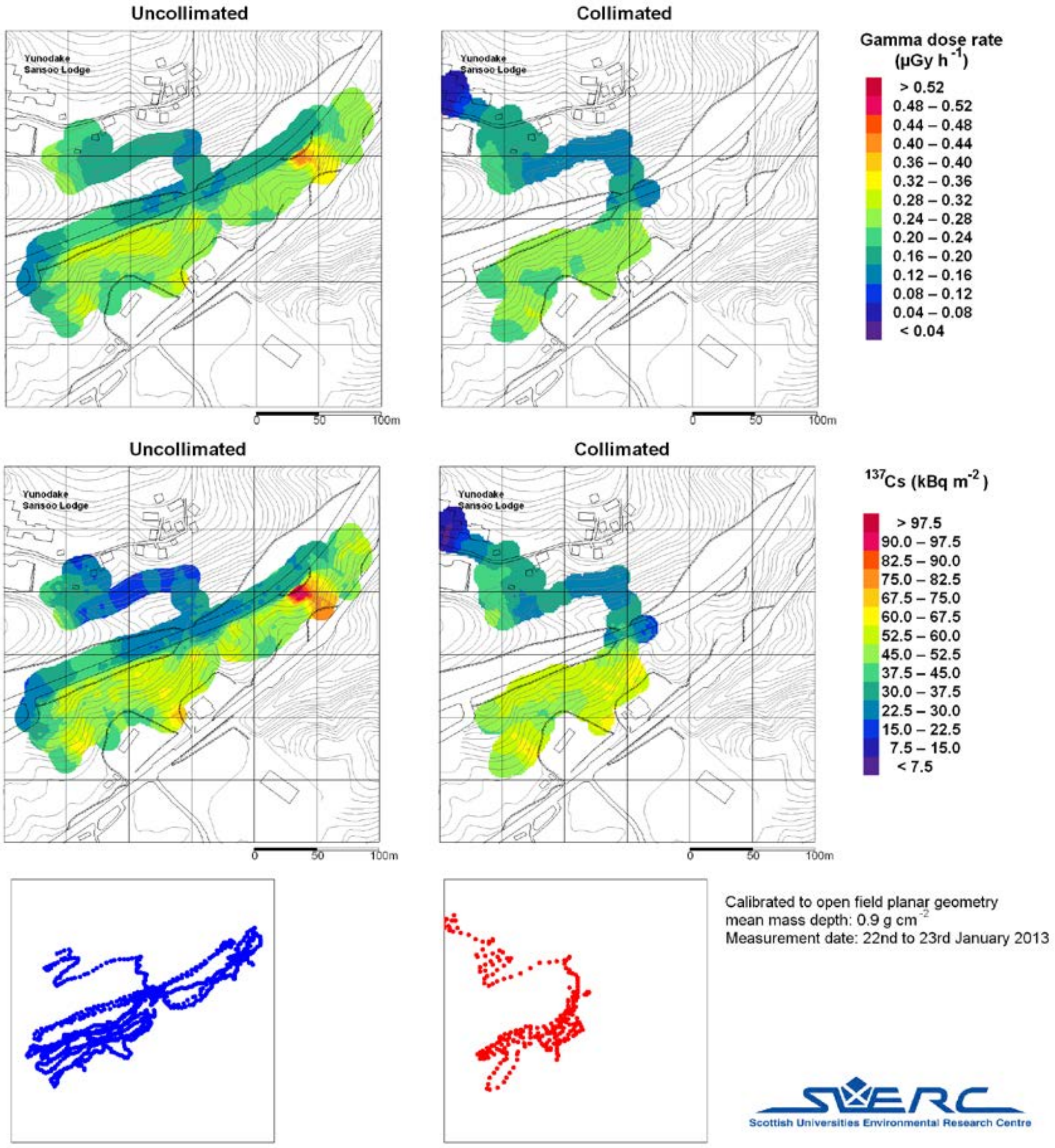


Figure 3: Dose rate (top) and ^{137}Cs activity per unit area (middle) measured in January 2013, with and without the collimator. The measurement positions are shown at the bottom.



Calibrated to open field planar geometry
mean mass depth: 0.9 g cm^{-2}
Measurement date: 22nd to 23rd January 2013

Figure 4: Snow depth in February 2014. Uncertainties in mass depth estimates are typically ±15 kg m^{-2}.

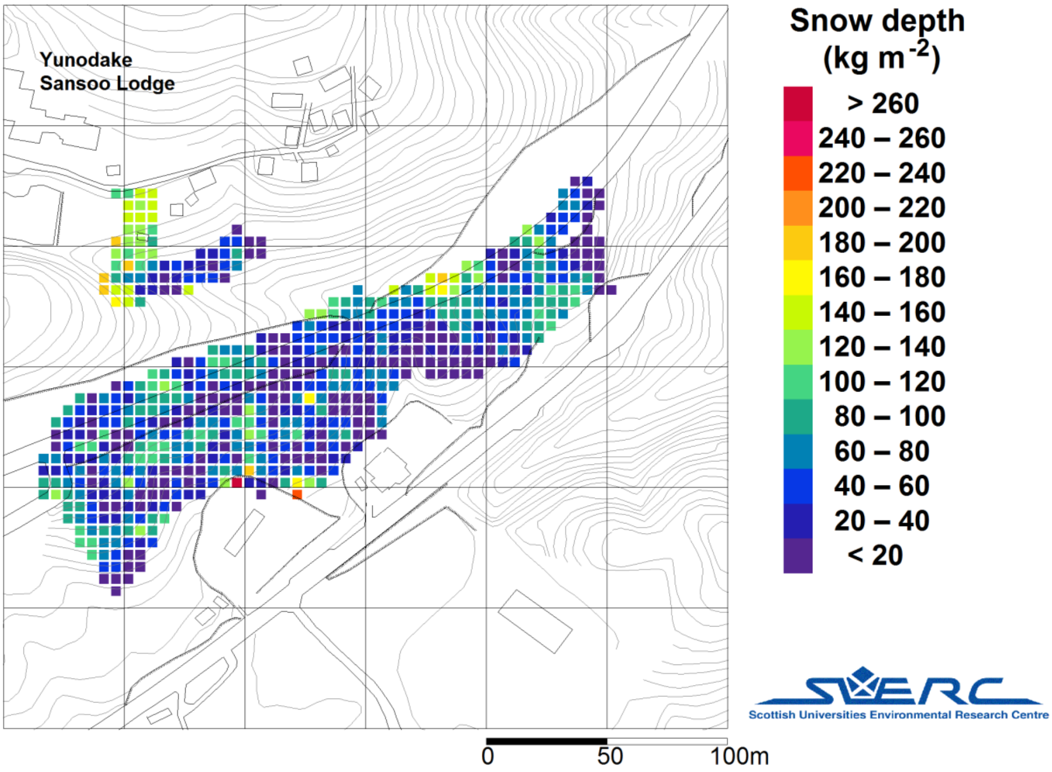


Figure 5: ^{137}Cs activity per unit area and dose rate in February 2014, following correction for snow attenuation. The remediated area (15m x 45 m) is indicated with a dotted boundary within the SW part of the survey area

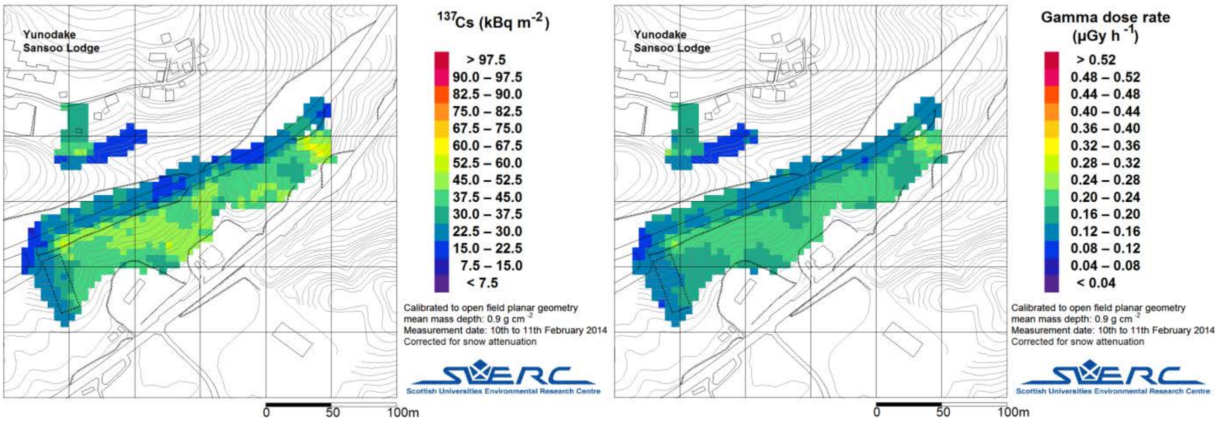


Figure 6: Ratios between 2014 and 2013 measurements for ^{137}Cs activity per unit area and dose rate, after accounting for physical decay and snow attenuation. Values greater than 1.0 indicate an increase in activity over the year, values less than this a decrease.

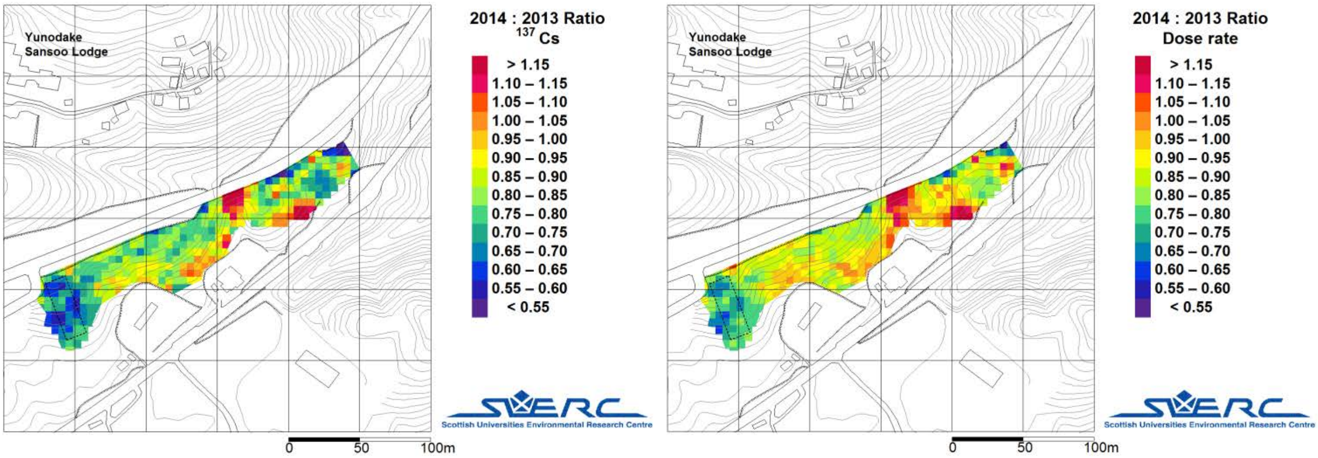


Table 1: Summary of surveys, showing the date, the systems used with measurement times and the number of measurements.

Date	Tasks	Number of measurements
22nd January 2013	System 1: no collimator (10 s)	480
	System 1: collimator (10 s)	160
	System 2: no collimator (5 s)	1270
	System 2: no collimator (10 s)	960
23rd January 2013	System 1: collimator (10 s)	200
	System 2: no collimator (10 s)	590
10th February 2014	System 2: no collimator (5 s)	1340
	System 3: no collimator (5 s)	900
	System 3: collimator (5 s)	390
11th February 2014	System 3: collimator (5 s)	940

Table 2: Mean and standard error of ^{134}Cs and ^{137}Cs activity per unit area and dose rate for the cedar forest measured with and without the collimator in 2013 and 2014, with the reductions in apparent activity and dose rate resulting from the use of the collimator. Data are restricted to the areas which were not remediated. No decay or snow attenuation corrections are applied.

Year	Survey	Radiocaesium (kBq m^{-2})		Dose rate (μGy h^{-1})
		^{134}Cs	^{137}Cs	
2013	Uncollimated (459 measurements)	25.8 ± 0.2	53.0 ± 0.4	0.258 ± 0.001
	Collimated (174 measurements)	24.7 ± 0.2	53.0 ± 0.4	0.253 ± 0.002
	Reduction	1.1 ± 0.3	0.0 ± 0.6	0.005 ± 0.002
2014	Uncollimated (636 measurements)	14.2 ± 0.1	30.6 ± 0.2	0.136 ± 0.001
	Collimated (225 measurements)	12.4 ± 0.1	30.3 ± 0.3	0.128 ± 0.001
	Reduction	1.8 ± 0.1	0.3 ± 0.4	0.008 ± 0.001

Table 3: Mean and standard error of ^{134}Cs and ^{137}Cs activity per unit area and dose rate for the remediated and control areas of the cedar forestry for 2013 and 2014. All data are decay corrected to February 2014, with snow attenuation accounted for.

		^{134}Cs (kBq m^{-2})	^{137}Cs (kBq m^{-2})	Dose rate (μGy h^{-1})
Remediated	2013	18.3 ± 0.4	42.0 ± 1.1	0.188 ± 0.003
(54 cells)	2014	13.4 ± 0.3	29.0 ± 0.5	0.142 ± 0.002
	Reduction	27 ± 3 %	31 ± 3 %	24 ± 2 %
Control	2013	22.1 ± 0.2	52.4 ± 0.6	0.224 ± 0.002
(113 cells)	2014	20.3 ± 0.2	43.3 ± 0.4	0.199 ± 0.002
	Reduction	7.9 ± 1.5 %	17.5 ± 1.3 %	11.0 ± 1.1 %