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1 Evaluation of Forest Decontamination Using Radiometric Measurements

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9

10 Abstract

11 An experiment has been conducted to evaluate the additional dose reduction by clear felling 12 contaminated forestry in Fukushima Prefecture, Japan, and using the timber to cover the areas 13 with wood chips. A portable gamma spectrometry system, comprising a backpack containing 14 a 3x3" NaI(Tl) detector with digital spectrometer and GPS receiver, has been used to map 15 dose rate and radionuclide activity concentrations before, after and at stages during this 16 experiment. The data show the effect of the different stages of the experiment on dose rate at 17 different locations around the site. The spectrometric data have allowed the assessment of the 18 contributions of natural and anthropogenic radionuclides to the dose rate at different parts of 19 the site before and after the experiment. This has clearly demonstrated the value of 20 radiometric methods in evaluating remediation, and the effect of other environmental 21 processes. The value of spectrometric methods which directly measure radionuclide 22 concentrations has also been shown, especially through the identification of the contribution 23 of natural and anthropogenic activity to the measured dose rate. The experiment has shown 24 that clearing trees and applying wood chips can reduce dose rates by 10-15% beyond that 25 achieved by just clearing the forest litter and natural redistribution of radiocaesium.

27 Keywords

28 Radiocaesium; Gamma Dose Rate; Fukushima Nuclear Accident

29

30 **1. Introduction**

31 Following the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in March 2011, 32 large areas of Fukushima Prefecture were contaminated with radiocaesium. Approximately 33 70% of the contaminated area is forested, providing a reservoir for contamination that is 34 largely recycled within the forests (Hashimoto et.al. 2013). Remediation of forests is 35 particularly challenging, and current practice in Japan is to reduce the dose rate in and around 36 buildings by the removal of forest litter and undergrowth to a distance of 20 m from roads 37 and buildings. Decontamination model projects conducted by the Japan Atomic Energy 38 Agency (JAEA) have demonstrated reductions in dose rates within remediated forest areas of 39 30-50% by such methods, with higher reductions achieved on open areas (Hardie & 40 McKinley 2014, Nakayama et.al. 2015). Similar remediation factors have been observed in 41 backpack surveys of a forest remediation trial, involving litter removal, near Iwaki 42 (Sanderson et.al. 2016).

Another potential method for forest remediation is to cut down trees near households. This has advantages since it eliminates the risk of future deposition through litter fall and through fall (Kato et.al. 2015). However, this approach has not yet been tested, and therefore there is no data available to assess the effect of forest removal on dose rate.

47 Radiometric surveys using backpacks are ideal to evaluate forest dose rates and the effect 48 of decontamination. Within forests dose is spatially highly heterogeneous, and the random 49 distribution of tree trunks make regular grid data measurements difficult. Backpack surveys 50 are capable of total area coverage, and can produce maps of the distribution of dose rate and radionuclide deposition. Repeat surveys can monitor the changes in the spatial distribution of
dose rate due to forest decontamination work.

53 In the work described here, an experiment has been conducted to assess the effect of 54 additional decontamination measures on the dose rate recorded at the edge of contaminated 55 forests. Trees along the edge of the forest, to a distance of 10 m, have been felled, and wood 56 chips derived from the felled trees applied to the ground surface. Assuming a density of approximately 500 kg m⁻³, 1-2 cm of wood chips over a near-infinite surface would be 57 expected to reduce radiocaesium full energy count rates by 10-20% and dose rates by 7-17%. 58 59 When applied to smaller areas the expected reductions will be smaller. At various stages 60 during the experiment portable gamma spectrometry methods were used to map the spatial 61 distribution of dose rate and deposited activity within and around the remediated area. These 62 data are used to assess the reduction in dose rate achieved at different points in the experiment. 63

64 **2.** Methodology

65 **2.1 Site description**

The experiment was conducted at the former Yamakiya Elementary School, Kawamata 66 67 town (37°36.169N, 140°40.582E), approximately 40 km from the FDNPP. This is within the 68 evacuated area, categorised as an area where preparations are underway to allow residents to 69 return to their homes. The location of the site is shown in Fig. 1. The site has two connected 70 buildings housing the elementary school and a separate building for a kindergarten, with a 71 sports field on a levelled terrace. A forest area, with a mixture of cedar, beech, Japanese oak 72 and red pine, is situated to the north and east of the school buildings. To the east, the forest is 73 dominated by deciduous trees and is approximately 10 m above the school buildings with a 74 grassed slope at approximately 45° to the kindergarten building and a small enclosed space. Beyond this, towards the south eastern part of the forest red pine are also present. To the 75

north, the forest coniferous trees are more numerous and descends northwards below the school terrace, with the edge of the forest descending until it is level with the elementary school building. The lower parts of this slope are dominated by red pine. The forest site is being used for ongoing studies of the behaviour of radiocaesium in forest environments, including studies of interception and transfer from the forest canopy (Kato & Onda 2014,

81 Kato et.al. 2015) and depth distribution in soils (Takahashi et.al. 2015).

Decontamination work was carried out at the school in 2012, following current practice for remediation of contaminated areas. Top soil was removed from the sports field and grassed areas around the school and replaced with sand or fresh turf between May and September 2012, and leaf litter removed from the forest areas around the school to a distance of 20 m from the forest edge between October and December 2012.

87 To facilitate comparisons between surveys, the site was divided into a series of areas (shown in Fig. 2); A to D for artificial surfaces around the school, Area C also includes a 88 89 small ornamental garden area outside the main entrance. E to H for the forest and grassed 90 areas. Area E was the section of forest that was felled during this work, and with area G had 91 previously been remediated by the removal of leaf litter. Areas E, F, and G have been divided 92 into three sub areas, and area H two sub areas. These areas are described in Table 1. Static 93 dose rate measurements were also collected at marked positions on two overlapping grids 94 (Fig. 2). Both grids had 10 m spacing, with one axis parallel to the edge of the forest to the 95 east of the school. On the northern part of the site, the forest edge runs north-south, with the 96 axes aligned along the western edge of area E2 and the southern edge of area E3. South of the 97 kindergarten building the forest edge turns slightly to the west, resulting in a rotational offset 98 between the grids, with the axis here aligned along the western edge of area E1.

99

100 **2.2 Experimental timescale**

101 Surveys were conducted prior to the start of the experiment, at intervals during the 102 experiment, and following completion of the experiment. Each survey included the area of 103 forest which was felled, and surrounding areas of forest and school grounds, but some of the 104 surveys did not include all of the sports field and areas around the school more distant from 105 the experimental area. Details of these measurements are given in Table 2. The trees to a distance of approximately 10 m into the forest were felled on the 8th, 9th (Areas E1 and part of 106 E2), 12th and 13th (part of Area E2 and Area E3) November 2014, with the trunks cut and 107 108 temporarily stacked on site at the edge of the remaining forest (see Fig. 3 (d)). The majority 109 of the tree trunks were subsequently chipped on site, with the chips applied to part of the 110 slope between the 23rd and 30th November (Area F1). Branches and other litter left on the 111 felled area (Area E) were mostly removed between the 15th and 23rd November, with 112 substantial stacks of branches remaining along the bottom of the slope to the east of the 113 school buildings (the southern end of Area F1) and Area D. Following an interruption in 114 work during the winter, the remaining area of slope (Area F2) was covered in wood chips, 115 and the majority of the remaining piles of branches removed on the 28th April 2015 with the 116 last few remaining material removed after that. Figure 3 shows photographs of the site at 117 different stages of the decontamination experiment.

118

119 **2.3 Instrumentation and data processing**

Measurements were conducted using a Portable Gamma Spectrometry System developed at the Scottish Universities Environmental Research Centre (SUERC). The system comprises 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 5 s for each spectrum, corresponding to averaging the signal over a distance of approximately 2 m. As far as possible within the

126 constraints of the terrain, a dense survey pattern of parallel lines 1-2 m apart was maintained. 127 A netbook or tablet computer was used to power the system, and run custom software that 128 continuously logged spectra with associated positional information, and run real-time 129 analysis using predetermined calibration parameters. Real-time data analysis used a spectral windows with stripping algorithm to calculate activity per unit area for ¹³⁷Cs and ¹³⁴Cs, and 130 activity per unit mass for ⁴⁰K, ²¹⁴Bi (²³⁸U decay series) and ²⁰⁸Tl (²³²Th decay series), and a 131 scaled count rate above 450 keV to calculate total terrestrial gamma dose rate. Analysis 132 133 includes subtraction of a background recorded from a plastic boat over open water, 134 accounting for internal activity within the detector and the cosmic dose rate. For the detector 135 used here, these measurements were conducted on Loch Lomond, Scotland (56°8N, 4°38W), 136 it is recognised that the sea level cosmic background is slightly higher for these 137 measurements than would be the case in Fukushima, but this is an insignificant error 138 compared to the terrestrial radiation. This method, applied to airborne measurements, has 139 been described in numerous places including IAEA (1991, 2003) and Sanderson et.al. (1994), 140 Cresswell et.al. (2006) also includes the derivation of uncertainties on measurements. 141 The calibration parameters for the real-time analysis, which take account of the shielding 142 effect of the operator (Buchanan et.al. 2016), were validated using reference sites in Scotland 143 and Japan (Cresswell et.al. 2013, Sanderson et.al. 2013). The parameters for radiocaesium 144 and natural activity concentrations assume an open field geometry, and do not account for 145 shielding effects from trees or contributions to the radiation measured from activity in the 146 canopy. Measurements in a cedar forest near Iwaki demonstrated that the contribution to the measured signal from activity in the canopy is no more than a few percent, even if the canopy 147 148 retains a significant fraction of the activity (Sanderson et.al. 2016). An assessment of the 149 shielding effects of trees and understory plants is ongoing, but initial analysis suggests that this results in a reduction in count rate of less than 20% compared to the open field for forests 150

of similar density to this site. The relative changes in activity concentration and dose rate in areas that are not physically changed during this experiment will not be affected by this attenuation effect. However, it may be necessary to consider this effect in comparing surveys of areas where trees were removed.

In open field conditions, the detector field of view is approximately 10-15 m radius,
depending on source depth profile (Tyler et.al. 1996). The effect of attenuation by trees will
be to reduce this field of view, to less than 10 m. This has not, however, been determined
precisely to date.

159 For natural series radionuclides, the calibration assumes a uniform depth distribution. For 160 radiocaesium, the calibration assumes an exponential depth distribution with a relaxation mass depth of 1.0 g cm⁻², which matches calibration sites established in Fukushima in 2012 161 (Sanderson et.al. 2013). Soil samples collected from the forest at Yamakiya between July 162 163 2011 and December 2012 (Takahashi et.al. 2015) show relaxation mass depths, excluding the litter layer, of between 0.4-0.9 g cm⁻². Including the litter layer and subsequent measurements 164 (see Table S.1, Supplementary Material) the range of mass depths is 0.3-0.4 g cm⁻². Three 165 soil cores collected in May 2016 at a different location within the school forest had mass 166 depths between 0.5 and 1.1 g cm⁻². Therefore, although it is likely that the mass depth across 167 the site may be less than the assumed mass depth of 1.0 g cm^{-2} , this is still appropriate for this 168 169 location. The uncertainty on mass depth results in a systematic error on the activity per unit 170 area of less than 20%, but has no impact on the relative values measured at different times 171 assuming no change in depth profile between measurements. Measurements at a reference 172 site on the Fukushima University just prior to field work at Yamakiya in November 2014, and 173 at the Fukushima Prefecture Fruit Tree Research Institute in April 2015, confirmed the 174 functionality of the detector and the validity of the calibration coefficients. These sites have been extensively sampled with activity per unit area and depth profiles determined by 175

176 laboratory gamma spectrometry relative to international reference materials (Sanderson et.al.177 2013).

178 The dose rate (μ Gy h⁻¹) and ¹³⁷Cs and ¹³⁴Cs activity per unit area (kBq m⁻²) and natural 179 series activity per unit mass (Bq kg⁻¹) have been mapped using a modified inverse distance 180 weighting algorithm, with the average value for each map pixel being given by:

$$\bar{A} = \frac{\sum_{i} w_i A_i}{\sum_{i} w_i}$$

181 where the summation is across all points *i* within a maximum range r_{max} of the map pixel. 182 The weight assigned to each point, w_i , is given by:

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

183 Where r_i is the distance between the measurement point and the map pixel, Γ is a constant 184 that flattens the distribution at small values of r_i , and p is a power. For this work, a power p=1.7, $\Gamma=1$ m and maximum range $r_{max}=10$ m have been used. The combination of power and 185 186 flattening constant results in 95% of the weight being carried by measurements within 4 m of 187 the pixel, approximately comparable to the field of view of the detector. The maximum range 188 allows two-three measurements in any direction to be included in the weighted mean value. 189 To allow comparisons between data collected on different occasions, a spatial regridding 190 algorithm is employed (Sanderson et.al 2004, 2008). This uses the modified inverse distance 191 weighting algorithm to determine values for dose rate, activity per unit area or activity per 192 unit mass in each of a grid of cells. For this work, this has been done using cells of 5x5 m, 193 and the same parameters for the interpolation as were used to generate the mapped data. 194 The surveys were conducted over an extended period in autumn and spring. There is the 195 potential for changing environmental conditions, in particular soil water content, to influence 196 the radiometric data. If it is assumed that the activity per unit dry mass for natural 197 radionuclides is constant then variations in the measured activity per unit mass would reflect changing soil water content. With the backpack system used here the ⁴⁰K peak count rate is 198

used, the peaks used for U-series (1764 keV 214 Bi) and Th-series (2614 keV 208 Tl) activity 199 measurement have lower intensities and are measured with lower efficiency, and hence carry 200 substantial statistical imprecision. Ratios for different parts of the survey area are produced 201 between the ⁴⁰K count rates for each survey to the first survey, with average ratios produced 202 203 for the areas with predominantly artificial surfaces and the forests. A relative soil density 204 compared to the initial survey conditions, from which a relative mean mass depth for radiocaesium can be determined. Interpolation of fluence rates for ¹³⁷Cs (662 keV) and ¹³⁴Cs 205 (795 keV) and dose rate conversion coefficients tabulated by ICRU (1994) as a function of 206 207 mass depth can then be used to adjust the measured activity per unit area for radiocaesium 208 and dose rates to values for the environmental conditions of the initial survey. 209 To better understand the origins of the dose rates observed, dose rate apportionment has 210 been used to determine the contribution of the dose rate from natural radionuclides and 211 radiocaesium. This uses conversion factors to determine air kerma dose rate from the activity 212 concentrations measured using the portable gamma spectrometry system, and the percentage 213 of the total dose rate from each of these sources. Measurements of dose rate (ambient dose, μ Sv h⁻¹) were also collected using a TCS-172B 214 survey meter (Aloka Hitachi Medical, Japan, calibrated by Chiyoda Technology Corporation, 215 Japan, on September 24th 2014) at a height of 1 m. During most of the surveys, the dose rates 216 217 determined with the backpack from 4-8 measurements while standing at some of these 218 positions were also recorded. 219

- 220 **3. Results and discussion**
- 221 **3.1 Detector response validation**

Soil layer samples were collected on four occasions between July 2011 and December 2012
from a location within the forest to the east of the school which had not been remediated

224 (Takahashi et.al. 2015), with three additional samples collected on an annual basis subsequently (see Table S.1, Supplementary Material) with a mean (± standard deviation on 225 seven measurements) activity per unit area for 137 Cs of 341 ± 85 kBq m⁻² and for 134 Cs of 100 226 \pm 19 kBq m⁻² (decay corrected to November 2014). The average activity per unit area 227 measured with the backpack system within 10 m of this location on the 8th November 2014, 228 prior to felling of trees, was 264 ± 67 and 89 ± 23 kBg m⁻² for ¹³⁷Cs and ¹³⁴Cs respectively. 229 230 Although this single point observation of non-contemporaneous measurements is not ideal, 231 the backpack and soil samples agree within measurement uncertainties. Together with the 232 extensive detector validation previously conducted this does indicate that the calibration 233 assumptions are valid for this forest environment, and suggests that the effects of attenuation 234 by the trees and radiation from the canopy are relatively small. 235 During the surveys with the portable gamma spectrometry system, dose rates were recorded

236 at some of the measurement points used to conduct dose rate measurements using the dose 237 rate meter. Table S.2 in the supplementary material gives the dose rates recorded at these 238 locations using the SUERC spectrometer, and measurements with the dose rate meter at 239 similar stages in the experiment. Figure 4 shows the measurements by the two instruments at 240 common points. The slope of a linear regression through these points reflects the approximate 241 relationship between the different dose rate units, 1 Gy \approx 1.2 Sv, with the small non-zero 242 intercept probably reflecting differences in assumed instrumental background. Again, these 243 observations support the prior validation of the backpack instrument.

244

245 **3.2 Environmental conditions**

As noted, there is evidence of variations in environmental conditions, probably soil water
content, affecting the activity concentrations and dose rates measured by the backpack
system. Average values for the activity concentrations recorded for different parts of the site

for each survey are given in Table S.3 in the supplementary material. Ratios of ⁴⁰K activity 249 250 per unit mass for each survey relative to the initial survey were used to quantify these 251 variations. It is observed that for the artificial surfaces (areas A-D) these ratios were not 252 significantly different from unity, and hence no corrections were required for these areas. However, within the forest (areas E-H) there were some significant variation in these ratios 253 254 for different surveys, but with less variation between different parts of the forest for each survey. An average value of this ratio for all the forest areas was therefore used for each 255 256 survey.

On the assumption that soil water content was the only variable, and that the ⁴⁰K activity 257 258 per unit mass was constant with depth, these ratios can be used to calculate relative soil 259 density changes, and hence relative mass depth changes for radiocaesium. Other factors may 260 also contribute to variability in dose rates, including atmospheric conditions and variability in 261 cosmic ray intensity. All the surveys were conducted in dry weather. It would be expected 262 that air humidity and cosmic ray intensity would affect both forest and artificial areas, and the 263 absence of an appreciable effect on the artificial surfaces suggests that these are not 264 significant contributions to the observed variations in natural activity count rates. Correction factors for the radiocaesium activity per unit area and dose rate have been calculated using 265 266 tabulated photon flux data and dose rate conversion coefficients (ICRU, 1994) for the mass depths thus estimated. The ⁴⁰K activity ratios, relative soil density and correction factors 267 determined are given in Table S.4 in the supplementary material, with the corrected values 268 269 also given in Table S.3.

For the surveys on the 14-15th November 2014 and 28th April 2015 the average ⁴⁰K activity per unit mass in the forest areas (areas E-H) was smaller than for the initial survey on the 7th-8th November 2014, suggesting that the ground was wetter at these times. Conversely, for the 23rd and 30th November 2014 and especially the 27th May 2015 the measured activity

concentrations were higher, suggesting the ground was drier. It is also noted that on the 15th November 2014 survey an enhancement in ²¹⁴Bi activity per unit mass is measured in all areas, without corresponding variations in the ⁴⁰K and ²⁰⁸Tl values, which suggests a radon related effect on that day. The impact on total dose rate is very small, and this radon effect has not been corrected for. The advantage of spectrometric measurements compared to dose rate instruments, with the use of natural activity to assess environmental conditions, is evident from these analyses.

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282

2 **3.3 Dose rate variation with time**

The average dose rate for each area around the site, following adjustments for 283 environmental variations and the decay of ¹³⁴Cs, have been plotted in Fig. 5. For the artificial 284 285 surfaces around the school the dose rate is approximately constant through the experimental 286 period, with decreases of approximately 15% for area A (sports field) and approximately 25% 287 for area B (at the far end of the school). These are the most exposed areas on the site, and as 288 such may be prone to enhanced weathering effects, resulting in greater redistribution of 289 activity. Areas C (in front of the main entrance) and D (nearest the forest) have slightly 290 higher dose rates, probably attributable to the proximity to vegetated areas, which are 291 approximately unchanged over the duration of the surveys.

The area of forest where trees were felled (E) shows a decrease in dose rate following the felling of trees, followed by a further reduction while tree branches are removed, with no significant additional dose rate reductions following the application of wood chips to the adjacent area (F). Overall, a reduction in dose rate of $15 \pm 5\%$ is observed in this area as a result of felling the trees. The area (F) between the felled forest and the school shows initial increases in dose rate with the felling of trees, especially to the northern and southern parts of the area where tree branches were stacked. This is most likely the result of the accumulation

299 of branches, which would carry some radiocaesium contamination, in this area. Clearing the 300 branches and application of wood chips reduces the dose rate to $9 \pm 4\%$ below the initial 301 values. For the forest area adjacent to the felled area (G) a dose rate reduction following the 302 felling of trees was observed, with an increase while this area was used to stack tree branches 303 prior to production of wood chips. After clearing these branches, an overall dose rate 304 reduction of $12 \pm 4\%$ is observed. Area H, the control area not directly affected by the 305 experiment, shows a large reduction in dose rate in the second part of November 2014, for 306 reasons that are unclear, but overall only a small dose rate reduction $(7 \pm 3\%)$ during the 307 course of the experiment for most of the area. A much larger reduction in dose rate is 308 observed for the northern part of this area (H2) in May 2015, which is also unexplained. 309 Dose rate apportionments for different parts of the site are shown in Fig. 6, for data 310 recorded at each stage in the experiment. Areas A, B, C and D all have similar natural series 311 activity concentrations, with natural dose rates of 0.050-0.055 μ Gy h⁻¹. Area A, the school 312 sports field, has the smallest contribution from radiocaesium to the total dose rate (65%). 313 Areas B, C and D contain some natural surfaces with small ornamental trees, with area D 314 adjacent to the forest area, and as a consequence have slightly higher radiocaesium concentrations from these harder to remediate surfaces (contributing 75-80% of the dose 315 316 rate).

At the conclusion of the experiment, the sports field (A) remains unchanged with the other areas around the school. (B, C and D) showing a small reduction in dose rate, an average of 5 \pm 3% beyond that from the physical decay of ¹³⁴Cs, as previously noted. In all the forest areas, the dose rate from natural series activity is approximately half that from the artificial surfaces of the school grounds (0.017-0.027 μ Gy h⁻¹), reflecting higher natural series activity concentrations in the construction materials of the school, and potentially the sand used to replace contaminated soil, compared to the local soils. Area F, the grassed slope between the

forest and the school, has natural dose rates between the two extremes reflecting
contributions from both environments. The variations in natural dose rates within each area
between surveys are within measurement uncertainties and demonstrate that the adjustments
for soil moisture content assuming constant natural series activity have removed variations
from this source. The reductions in the contributions from radioacaesium follow the same
pattern as previously noted for the total dose rate.

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331 **3.5 Dose rate and ¹³⁷Cs distribution at different stages during the experiment**

Maps of dose rate and ¹³⁷Cs activity per unit area for the different stages of the experiment are shown in Figs. 7 and 8. The ¹³⁴Cs distributions match the ¹³⁷Cs distributions and the corresponding maps are not reproduced here. These maps include adjustments to account for the physical decay of ¹³⁴Cs and changing environmental conditions.

336 The maps prior to felling trees (Figs 7(a) and 8(a)) clearly show the effect of earlier remediation, with the school sports fields showing dose rates below $0.15 \,\mu\text{Gy}\,\text{h}^{-1}$ and ^{137}Cs 337 below 50 kBq m⁻², other areas around the school showing dose rates below 0.40 μ Gy h⁻¹ and 338 137 Cs below 150 kBq m⁻², and the remediated areas of the forest showing dose rates below 339 0.70μ Gy h⁻¹ and ¹³⁷Cs below 300 kBg m⁻². In contrast areas of the forest that have not been 340 remediated show dose rates greater than 0.90 μ Gy h⁻¹ and ¹³⁷Cs greater than 300 kBq m⁻². 341 342 Comparison between the forest areas which had been remediated in 2012 and adjacent unremediated forest suggests a reduction in dose rate following remediation of approximately 343 344 35-60%, consistent with earlier studies on other sites (Nakayama et.al. 2015, Sanderson et.al. 345 2016).

An average dose rate of $0.1 \,\mu\text{Gy}\,\text{h}^{-1}$ would result in an annual dose of approximately 1mSv. Current policy in Japan is to set a target level for protective measures in the lower part of the 1-20 mSv y⁻¹ range (NSC 2011). Thus the remediated sports fields are already at this target

level, but the remediated forest areas are still at the higher end of the range for the policytarget.

The areas which had not been remediated in 2012 show similar values for the ¹³⁷Cs activity 351 352 per unit area, despite variation in dominant tree type, which is consistent with previous observations at the site which showed no difference in uncalibrated ¹³⁷Cs count rates at 353 354 ground level for deciduous and coniferous trees (Kato & Onda 2014). However, in other locations in Japan significantly elevated deposition has been observed for coniferous trees 355 356 compared to deciduous (Yoshihara et.al. 2013, Sanderson et.al. 2016). This suggests local 357 variation in the importance of foliar interception, with factors including the relative 358 contributions of wet and dry deposition and stand density contributing. 359 In the areas where leaf litter was removed in 2012 there are differences observed in 137 Cs 360 activity per unit area. In areas dominated by deciduous trees, predominantly to the east of the 361 school, activity per unit area in the remediated forest ranges from approximately 100-200 kBq m^{-2} . Where coniferous trees dominate, activity per unit area is higher, between 150-250 362 kBq m⁻². It is suggested that this reflects differences in interception and subsequent transport 363 364 of radiocaesium. Conifers intercepted radiocaesium in the canopy, with transfer to needles 365 which fall to the litter layer over several years. In March 2011 the deciduous trees would not 366 have had leaves, and consequently interception of radiocaesium in the canopy would be much 367 lower and a larger proportion would be deposited directly onto the ground surface. Therefore, 368 when the litter layer was removed during 2012 a larger proportion of the activity was 369 removed from the deciduous areas. Subsequent build up of litter would have included 370 contaminated needles from the conifers with much lower concentrations of radiocaesium in 371 the leaves of the deciduous trees.

The maps for data recorded during the experiment (Figs. 7(b)-(e) and 8(b)-(e)) show no significant redistribution of activity over the majority of the area. The ornamental area by the

374 school entrance becomes increasingly evident over this period. A small enhancement in dose rate to the south of the kindergarten on the 14th-15th November (Figs. 7(b) and 8(b)) 375 corresponds to an area used to stack branches removed from the trees that had been cut down. 376 377 The final survey concentrated on measurements of the areas that had previously been used 378 as temporary stacks for branches and other tree material (predominantly in area D and F1), 379 and the density of observations in the forest (area H1) is significantly lower than on the previous surveys. This has resulted in the stripes evident in Figs. 7(f) and 8(f), in particular 380 for ¹³⁷Cs, which are an artefact of the increased survey line spacing and are not considered to 381 382 represent a difference in depositional pattern. For the area to the east of the buildings where trees were felled (areas E1 and E2), the ¹³⁷Cs activity per unit area at the end of the 383 384 experiment is very similar to the initial state. To the north of the buildings (area E3), the corresponding area where trees were felled has a slightly higher ¹³⁷Cs activity per unit area. 385 The data collected on the 28th April 2015 did not show as pronounced a soil water effect, and 386 for these data the area to the east of the buildings (E1 and E2) shows a small decrease in 137 Cs 387 388 activity per unit area with no significant change for the area to the north (E3). It appears that the effect of tree felling in reducing the ¹³⁷Cs inventory is different for these two areas, with it 389 390 being more effective to the east of the buildings than the north.

391 Figure 9 shows the change in dose rate for the areas of main interest for this study, after 392 regridding to allow for corrections due to environmental change and the physical decay of 134 Cs. The uncertainties propagated through the analysis result in typically 5-10% uncertainty 393 394 on this change, corresponding to one or two colour divisions, with uncertainties being larger 395 where the measurement density is lower and near the edges of the survey areas. The 396 combination of measurement uncertainties and local redistribution of activity, over ranges of 397 a few meters, results in a complicated visual pattern. However, some general observations 398 can still be made from these maps of dose rate change. There are small reductions in dose rate

399 observed with the felling of the trees (area E) particularly evident in Fig. 9(c), accompanied 400 by temporary increases in dose rate in locations where cut branches were stacked prior to 401 removal (to the north and south of the kindergarten, parts of areas D and F1) in Figs. 9(a)-402 9(c). Removal of the litter generated during the felling, and accumulated since the 2012 403 decontamination, had a very small impact on dose rate with the exception of locations where 404 this included removal of some of the stored branches, seen in comparisons of area D in Figs 405 9(a)-9(c) compared to 9(d). The area to which wood chips had been applied (to the south and 406 east of the kindergarten, areas F1 and F2) shows a small further reduction in dose rate in Fig. 407 9(e).

408

409 **4.** Conclusions

410 The various measurements presented here all support the conclusion that, on this site, 411 felling trees coupled with the use of wood chips to cover the ground has produced reductions 412 of 15±5% in the dose rate for the area which was cleared, and slightly smaller dose rate 413 reductions in adjacent areas both within the forest $(12\pm4\%)$ and in the open areas adjacent to 414 the forest (9 \pm 4%). However, it also noted that 7 \pm 3% reductions are measured in some of the areas of forest which were not subject to remediation, after consideration of the decay of 415 416 ¹³⁴Cs. Radiometric surveys on a cedar plantation near Iwaki have demonstrated reductions in 417 dose rate of 10-15% over a year in some unremediated areas (Sanderson et.al. 2016), which is 418 not dissimilar to the reductions observed here. If similar reductions in dose rate would have 419 occurred in the remediated areas as a result of natural redistributive processes then this 420 reduces the overall effectiveness of the remediation conducted in these areas to 421 approximately 10-15% reductions in dose rate. The reductions in dose rate that might be 422 achieved on other sites are likely to be variable, with dependencies on the particular characteristics of each site, including tree species and topography. 423

Although the felling of trees has a substantial impact on forest ecology, this does have an appreciable immediate impact on dose rates near the forest edge which will contribute to overall dose rate reductions in contaminated areas and ultimately to allowing evacuated residents to return to their homes. It is expected that the removal of trees, and hence a source of contaminated litterfall, will result in additional long term radiological benefits by reducing routes for recontamination of the remediated area. The Yamakiya study site will continue to be monitored over the next few years to evaluate this.

431 The dosimetry method using the convenient and sophisticated backpack system used in this432 work is widely applicable to decontamination work, and other studies, in Fukushima.

433

434 Supplementary Material

Table S.1: Data from scraper plate measurements at the Yamakiya school forest, July 2011 to
2015, and three cores collected in May 2016 from a second location within the forest.

437 Table S.2: Spot measurements (μ Gy h⁻¹ with the backpack, μ Sv h⁻¹ with the TCS-172B dose 438 rate meter) at the marked dose rate measurement points

439 Table S.3: Mean values for ¹³⁴Cs and ¹³⁷Cs activity per unit area, ⁴⁰K, ²¹⁴Bi and ²⁰⁸Tl activity

440 per unit mass and dose rate measured with the backpack system for different areas of the

441 Yamakiya site. Values in italics have been adjusted to correct for variable environmental

442 conditions and with 134 Cs decay corrected to 8^{th} November 2014.

Table S.4: Ratio of the mean ⁴⁰K count rates for each survey relative to the initial survey on the 7-8th November 2014, for the hard surface areas and the forest areas. For the forest area the relative soil density is given, and correction factors for both radiocaesium and dose rate calculated from this relative soil density difference.

447

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459

- 462 Buchanan, E.; Cresswell, A.J.; Seitz, B.; Sanderson, D.C.W. Operator related attenuation
- 463 effects in radiometric surveys. *Radiation Measurements* **2016**, 86, 24-31; doi
- 464 10.1016/j.radmeas.2015.15.029.

466 Cresswell, A.J.; Sanderson, D.C.W.; Harrold, M.; Kirley, B.; Mitchell, C.; Weir, A.

467 Demonstration of lightweight gamma spectrometry systems in urban environments. *Journal*

468 *of Environmental Radioactivity* **2013**, *124*, 22-28; DOI 10.1016/j.jenvrad.2013.03.006

469

- 470 Cresswell, A.J.; Sanderson, D.C.W.; White, D.C. 137Cs measurement uncertainties and
- 471 detection limits for airborne gamma spectrometry (AGS) data analysed using a spectral

472 windows method. Applied Radiation and Isotopes 2006, 64, 247-253; DOI

- 473 10.1016/j.apradiso.2005.07.013
- 474
- Hardie, S. M. L.; McKinley, I. G. Fukushima remediation: status and overview of future
 plans. *Journal of Environmental Radioactivity* 2014, *133*, 75-85; DOI
- 477 10.1016/j.jenvrad.2013.08.002

478

479	Hashimoto, S.; Matsuura	, T.; Nanko, K.; Linko [,]	v, I.; Shaw,	G.; Kaneko, S	. Predicted sp	oatio-
-----	-------------------------	-------------------------------------	--------------	---------------	----------------	--------

480 temporal dynamics of radiocesium deposited onto forests following the Fukushima nuclear

481 accident. *Scientific Reports* **2013**, *3*, srep02564. DOI: 10.1038/srep02564.

482

- 483 *Airborne Gamma Ray Spectrometer Surveying*; International Atomic Energy Agency:
- 484 Vienna, 1991; Technical Reports Series 323.

486	Guidelines for Radioelement Mapping Using Gamma Ray Spectrometry Data. International
487	Atomic Energy Agency: Vienna, 2003; IAEA-TECDOC-1363.

- 489 *Gamma-ray Spectrometry in the Environment*. International Commission on Radiation
- 490 Units and Measurements: Bethesda, 1994. ICRU Report No. 53. ISBN 0-913394-52-1
- 491

492 Kato, H.; Onda, Y. Temporal changes in the transfer of accidentally released 137Cs from

493 tree crowns to the forest floor after the Fukushima Daiichi Nuclear Power Plant accident.

494 *Progress in Nuclear Science and Technology* **2014**, *4*, 18-22.

- 495
- 496 Kato, H., Onda, Y., Hisadome, K., Loffredo, N., Kawamori, A. Temporal changes in

497 radiocesium deposition in various forest stands following the Fukushima Dai-ichi Nuclear

498 Power Plant accident. *Journal of Environmental Radioactivity* 2015.

499 doi:10.1016/j.jenvrad.2015.04.016

- 500
- 501 Nakayama, S.; Kawase, K.; Hardie, S.; Yashio, S.; Iijima, K.; Mckinley, I.; Miyahara, K.;
- 502 Klein, L. Remediation of Contaminated Areas in the Aftermath of the Accident at the
- 503 Fukushima Daiichi Nuclear Power Station: Overview, Analysis and Lessons Learned. Part 1:
- 504 A Report on the "Decontamination Pilot Project" Japan Atomic Energy Agency, 2015;
- 505 JAEA-Review 2014-051. doi: 10.11484/jaea-review-2014-051
- 506
- 507 Basic Policy of the Nuclear Safety Commission of Japan on Radiation Protection for
- 508 *Termination of Evacuation and Reconstruction*. Nuclear Safety Commission of Japan, 2011;
- 509 54th Nuclear Safety Commission Reference No. 4.
- 510 http://www.nsr.go.jp/archive/nsc/NSCenglish/geje/20110719suggest_4.pdf

512	Sanderson, D.C.W.; Cresswell, A.J.; Tamura, K.; Iwasaka, T.; Matsuzaki, K. Evaluating
513	remediation of radionuclide contaminated forest near Iwaki, Japan, using radiometric
514	methods. Journal of Environmental Radioactivity 2016, 162-163, 118-128. doi:
515	10.1016/j.jenvrad.2016.05.019
516	
517	Sanderson, D.C.W.; Cresswell, A.J.; Seitz, B.; Yamaguchi, K.; Takase, T.; Kawatsu, K.;
518	Suzuki, C.; Sasaki, M. Validated Radiometric Mapping in 2012 of Areas in Japan Affected by
519	the Fukushima-Daiichi Nuclear Accident. University of Glasgow: Glasgow, UK, 2013; ISBN
520	978-0-85261-937-7; http://eprints.gla.ac.uk/86365/
521	
522	Sanderson, D.C.W.; Cresswell, A.J.; White D.C. The effect of flight line spacing on
523	radioactivity inventory and spatial feature characteristics of airborne gamma-ray
524	spectrometry data. International Journal of Remote Sensing 2008, 29, 31-46; DOI
525	10.1080/01431160701268970
526	
527	Sanderson, D.C.W.; Cresswell, A.J.; Scott, E.M.; Lang, J.J. Demonstrating the European
528	capability for airborne gamma spectrometry: results from the ECCOMAGS exercise.
529	Radiation Protection Dosimetry 2004, 109, 119-125; DOI 10.1093/rpd/nch243
530	
531	Sanderson, D. C. W.; Allyson, J. D.; Tyler A. N. Rapid quantification and mapping of
532	radiometric data for anthropogenic and technically enhanced natural nuclides. In Application
533	of Uranium Exploration Data and Techniques in Environmental Studies; IAEA: Vienna
534	1994; IAEA TECDOC-827. pp 197-216.
535	

- 536 Takahashi, J.; Tamura, K.; Suda, T.; Matsumura, R.; Onda, Y. Vertical distribution and
- 537 temporal changes of 137Cs in soil profiles under various land uses after the Fukushima Dai-
- 538 ichi Nuclear Power Plant accident. Journal of Environmental Radioactivity 2015, 139, 351-
- 539 361; DOI 10.1016/j.jenvrad.2014.07.004
- 540
- 541 Tyler, A.N., Sanderson, D.C.W., Scott, E.M., Allyson, J.D. Accounting for Spatial
- 542 Variability and Fields of View in Environmental Gamma Ray Spectrometry. Journal of
- 543 Environmental Radioactivity **1996**, *33*, 213-235.
- 544
- 545 Yoshihara, T.; Matsumura, H.; Hashida, S.; Nagaoka, T. Radiocesium contaminations of 20
- 546 wood species and the corresponding gamma-ray dose rates around the canopies at 5 months
- 547 after the Fukushima nuclear power plant accident. Journal of Environmental Radioactivity

548 **2013**, *115*, 60-68. doi: 10.1016/j.jenvrad.2012.07.002

- 549
- 550





Figure 1. Location of the former Yamakiya Elementary School and forest (top), and the
buildings within the school (bottom). The location of the Takahashi et.al. (2015) sampling
site is indicated by the blue star, just to the south of the monitoring tower (Kato and Onda
2014, Kato et.al. 2015) indicated by the blue square. Aerial photograph © 2015 Google.
Image © 2015 ZENRIN.



- **Figure 2.** Location of survey area divisions (top), and dose rate measurement grids (bottom).
- 558 Aerial photograph © 2015 Google. Image © 2015 ZENRIN.



Figure 3. Photographs illustrating the different stages of remediation. (a) area E1 before
felling of trees, (b)-(c) areas E1 and E2 during tree felling, (d) area E1 after felling of trees,
(e) production of wood chips in area E2, (f) area F1 and (g) area F2 after production of wood
chips, (h) after application of wood chips on part of area F1, and (i) after application of wood
chips to whole of areas F1 and F2.



Figure 4. Comparison between dose rates measured with a survey meter and the backpack

567 system.



Measurement date
Figure 5. Variation in dose rate measured with the backpack system, after adjustments for
environmental variations and ¹³⁴Cs decay, with time for the artificial surfaces (A-D, top left),
the forest area that had been felled (E, middle left), the slope between the felled forest and the
school (F, top right), the unfelled forest previously remediated (G, middle right) and the rest
of the forest (H, bottom). Dose rate measurements with the TCS-172B survey meter are
shown (bottom left)



Figure 6. Dose rate apportionments at different parts of the site, as shown in Fig. 2 and listed

- 580 in Table 1, at each stage of the experiment. The areas that have been sub-divided in Fig. 2
- 581 have been combined here.



(b) 14th-15th November 2014, (c) 23rd November 2014, (d) 30th November 2014, (e) 28th April

2015 and (f) 27th May 2015.



Figure 8. ¹³⁷Cs activity per unit area distribution during the experiment. (a) 7th-8th November 2014, (b) 14th-15th November 2014, (c) 23rd November 2014, (d) 30th November 2014, (e)

- 589 28th April 2015 and (f) 27th May 2015.
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- 591



Figure 9. Dose rate changes relative to the initial survey (7-8th November 2014), with oranges and reds indicating an increase in dose rate and greens and blues a decrease, following corrections for environmental conditions and physical decay. Uncertainties are typically 5-10% (one or two colour bands) (a) Following removal of trees (14-15th November 2014), (b) following removal of litter (23rd November 2014), (c) following application of wood chips to part of the site (30th November 2014), (d) following completion of application

- 599 of wood chips (28th April 2015) and (e) following removal of the last of the cut trees (27th
- 600 May 2015). The subdivided areas (Fig.2 and Table 1) are indicated by the grey lines, and
- 601 labelled (f).

Area	Description	
А	Sports field south of the school buildings	
В	Footpath and line of trees to the north and west of the two parts of the Elementary	
	School building	
С	Area outside the Elementary School entrance, between the school and kindergarten,	
	including an ornamental shrubbery	
D	Open area to the north of the kindergarten and east of the Elementary school	
	buildings	
E	10 m wide forested area along the top of the slope east of the school buildings and to	
	the north of area D. This is subdivided into three sub-areas; E1 and E2 to the east of	
	the school buildings, E3 to the north. This area had previously been remediated by	
	removal of leaf litter.	
	Trees in this area were felled for this experiment.	
F	Slope between the forest and the school buildings, which levels off at the western	
	end of the northern section. This is subdivided into three sub-areas; F1 and F2 to the	
	east of the school buildings, F3 to the north.	
	Wood chips were applied to sub-areas E1 and E2 for this experiment.	
G	10 m wide strip of forest to the east and north of area E. This is subdivided into three	
	sub-areas; G1 and G2 to the east of the school buildings, G3 to the north.	
	This area had previously been remediated with the removal of leaf litter.	
Н	The remaining forestry around the school. This is subdivided into two sub-areas; H1	
	to the east and H2 to the north. Sub-area H1 is mixed woodland, sub-area H2	
	includes stands of red pine.	
T 11 4 D		

Table 1: Description of survey area divisions shown in Fig. 2.

Date	Stage	Area surveyed	Approximate number of
			measurements
7 th -8 th Nov	Prior to tree felling	School grounds, all forest to	1500
2014		the east, parts of forest to the	
		north including all areas	
		previously remediated	
14 th -15 th Nov	After tree felling	School grounds excluding	1200
2014		playground, forest 10-20 m	
		beyond felled area	
23 rd Nov	Following removal of	School grounds excluding	700
2014	litter	playground, forest 10-20 m	
		beyond felled area	
30 th Nov	Following application of	School grounds within 50 m of	700
2014	wood chips to part of the	forest, forest 10-20 m beyond	
	area	felled area	
28 th Apr	Following removal of cut	School grounds, all forest to	1800
2015	wood around school and	the east, parts of forest to the	
	application of wood chips	north including all areas	
	to entire area	previously remediated	
27 th May	Following removal off all	School grounds within 50 m of	1000
2015	branches and other	forest, all forest areas	
	material		

Table 2. Summary of surveys conducted.