森林立地55(2), 105～111 2013
Jpn J For Environ 55 (2), 105–111, 2013

Survey of radioactive contamination of sugi (*Cryptomeria japonica* D. Don) shoots and male flowers in Fukushima prefecture

Akio Akama[1,*], Yoshiyuki Kiyono[1], Tatsuo Kanazashi[1], Koji Shichi[1]
[1]Forestry and Forest Products Research Institute

We measured concentrations of radiocesium (^{134}Cs and ^{137}Cs) in shoots and male flowers of sugi (*Cryptomeria japonica* D. Don), sampled in Fukushima prefecture in 2011 and 2012, after the Fukushima Dai-ichi Nuclear Power Plant accident. The radioactivities of male flowers and shoots were mainly logarithmic linear to the air dose rates and radiocesium depositions. The measured radioactivities of male flowers ranged between 260 kBq kg^{-1} and < 0.1 kBq kg^{-1} in 2011. In 2012, average radioactivity decreased in the order old needles, one-year-old needles, male flowers, and current-year needles. Some radiocesium was probably absorbed by other organs and translocated within the tree. However, the average radioactivity of male flowers in 2012 was about 40% of the average value for the previous year. Thus, radiocesium absorption by sugi was apparently considerable for a short time after the accident, and subsequent absorption was lower.
Keywords : Fukushima prefecture, radiocesium, *Cryptomeria japonica* D. Don, male flower, air dose rate

赤間亮夫，清野嘉之，金指達郎，志知幸治：福島県内のスギの新梢と雄花における放射能汚染調査．森林立地55：105–111，2013.
東京電力福島第一原子力発電所事故後の2011年と2012年の秋に福島県内の広域においてスギの新梢と雄花を採取し，そこに含まれる放射性セシウム（^{134}Csと^{137}Cs)濃度を測定した。雄花と新梢の放射能は採取地の空間線量率および放射性セシウム沈着量と強い相関を示した。2011年秋の雄花の放射能は0.1 kBq kg^{-1}以下から260 kBq kg^{-1}の範囲であった。2012年秋における部位別の放射能は，2010年以前に展開していた旧葉で最も高く，2011年に展開した１年葉，雄花，2012年に展開した当年葉の順に低下した。旧葉または根から吸収されていた放射性セシウムが樹体内を転流し，雄花などに存在していたと考えられる。しかし，2012年の雄花における放射性セシウム濃度は前年の40％ほどに低下していた。これまでのところ，スギは事故直後に放射性セシウムをかなり吸収したものの，その後の吸収は少ないと考えられる。
キーワード：福島県，放射性セシウム，スギ，雄花，空間線量率

1. Introduction

The Fukushima Dai-ichi Nuclear Power Plant accident in March 2011 contaminated wide areas of eastern Japan with radioactive fallout, especially Fukushima prefecture. Approximately 70% of the land area of Fukushima prefecture is covered by forest, primarily sugi (*Cryptomeria japonica* D. Don) timber plantations. For these contaminated sugi plantations, it is most important to figure out the actual status of the radiocesium contamination and predict the dynamics of radiocesium within a tree and in the forest ecosystem.

Koarashi *et al.* (2012) observed much radiocesium in the litter layer at the forest sites, and Matsunaga *et al.* (2013) revealed low mobility of radiocesium in the soil. Kato *et al.* (2012) observed much radiocesium remained in the canopy after 5 months of the initial fallout. From these observations, Hashimoto *et al.* (2013) predicted the migration of radiocesium in the forest ecosystems.

Kuroda *et al.* (2013) revealed that the radiocesium concentrations in sugi timber samples taken from five sites rose with the density of radiocesium accumulation on the ground surface. However, it is not clear whether the radiocesium in the wood was absorbed from soil via roots or from aboveground parts directly contaminated by the fallout.

New shoots of sugi start to flush in May, and male flowers develop from July to October. Matured male flowers are dormant from November to February, and then pollen is dispersed from male flowers in spring. Therefore, newly flushed shoots and male flowers of sugi were not exposed to the radiocesium fallout at the time of the accident in March. However, Goor *et al.* (2007), Thiry *et al.* (2009) and Zhiyanski *et al.* (2010) investigated the root uptake of radiocesium by woody plants after the Chernobyl accident. In the case of Fukushima, Tagami *et al.* (2012) and Yoshihara *et al.* (2013) investigated radiocesium contaminations of wood species and demonstrated the translocation of radiocesium within the trees. Hirono and Nonaka (2012) reported that new buds of tea (*Camellia sinensis* (L.) O. Kuntze) in 2011 contained radiocesium even though bud flush was not until May. Thus, it is possible that new shoots of sugi, which is an evergreen tree like tea, might have been contaminated by

＊ 連絡・別刷請求先(Corresponding Author)：〒305–8687 茨城県つくば市松の里１ 森林総合研究所：Forestry and Forest Products Research Institute, 1 Matsunosato, Tsukuba, Ibaraki 305–8687, Japan
E-mail : akama@affrc.go.jp
1 独立行政法人森林総合研究所

(Received 30 July 2013, accepted 04 October 2013)

radiocesium. In addition, male sugi flowers develop near the tip of branches, and both male flowers and young shoots would likely accumulate radiocesium translocated within sugi trees. Furthermore, continuous absorption of radiocesium from soil would cause the radioactivity of wood to increase.

These characteristics of sugi suggest that sugi pollen dispersal might accelerate the diffusion of radiocesium from forests to suburbs. Therefore, in this study, we surveyed radiocesium concentrations in male flowers and shoots in sugi over a wide area of Fukushima prefecture, and clarified the state of radioactive contamination of sugi trees directly after the accident.

2. Materials and methods

We collected male sugi flowers from 131 sugi plantations in Fukushima prefecture in November or December 2011, and we collected male flowers and shoots from 24 of these 131 sugi plantations in November or December 2012. The 24 plantations were selected in 2012 to cover the entire range of air dose rate variation in 131 plantations with almost regular intervals. Three branches at the height of several meters in the sun were collected from different trees in each plantation. The same trees were not necessarily sampled in 2011 and 2012. Samples of three branches in each plantation at each sampling period were mixed. The air dose rates (μSv h^{-1}) of the plantations were measured simultaneously with the sampling in 2011 and 2012.

After sampling, male flowers were separated from branches. The shoots collected in 2012 were divided into current-year needles (developed in 2012), one-year-old needles (developed in 2011) and old needles (developed before the accident). All samples were immediately washed with tap water, rinsed with distilled water, oven-dried, and ground to powder with a cutting mill.

The processed male flower and needle samples were packed separately into 100-mL polystyrene containers for gamma-ray spectrometry measurements. The radiocesium activities of the samples collected in 2011 were measured by the Kyushu Environmental Evaluation Association (KEEA, Fukuoka, Japan), and those of the samples collected in 2012 were measured with an HPGe coaxial detector system (GEM20−70, Seiko EG & G, Tokyo, Japan) at Forestry and Forest Products Research Institute (FFPRI). Measurements were carried out for 1800 seconds or longer, and gamma-ray peaks of 604 and 662 keV were used to determine ^{134}Cs and

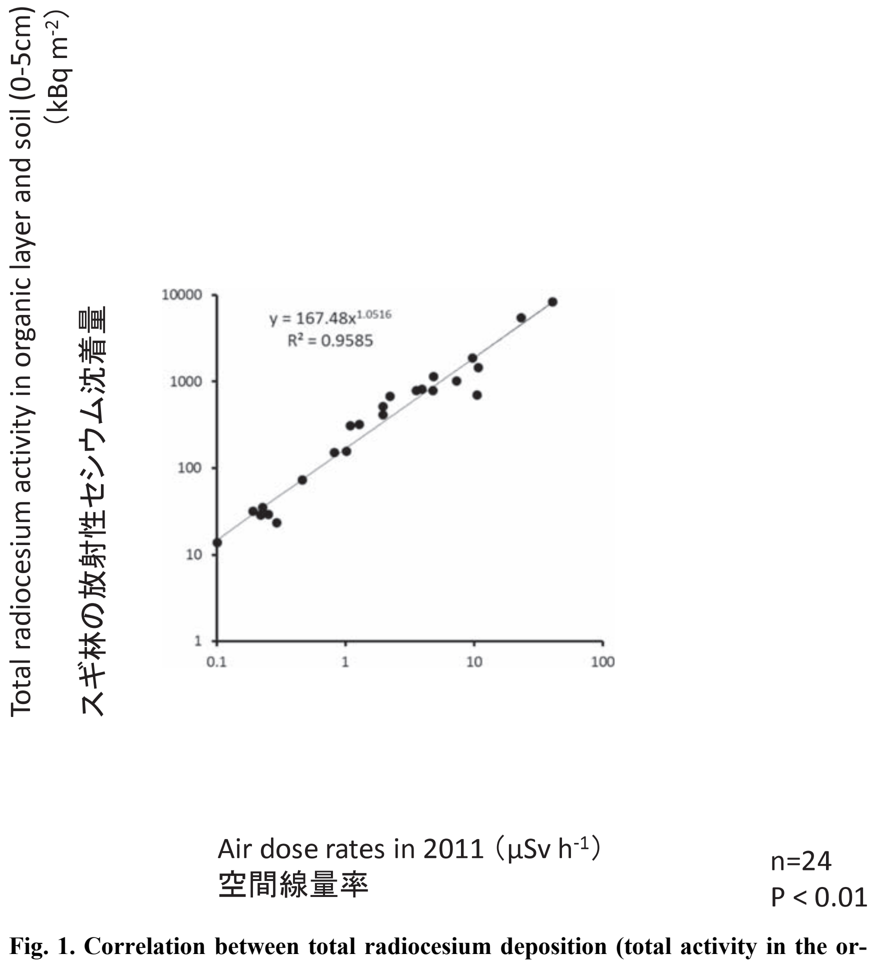

Fig. 1. Correlation between total radiocesium deposition (total activity in the organic layer and surface soil (0−5 cm)) and air dose rates in 2011

図−1. スギ林の放射性セシウム沈着量(堆積有機物層と土壌(0−5cm)の合計)と空間線量率の関係(2011年)

^{137}Cs activities, respectively. The measurement system was calibrated using a standard gamma-ray source (MX033MR, Japan Radioisotope Association, Tokyo, Japan). The measured ^{134}Cs activity was corrected for the sum effect of gamma rays. We found no differences in the measured activities in some samples that were measured at both KEEA and FFPRI. The decay correction was made for1st December in each sampling year. In our analysis in this paper, we use the total radiocesium activity concentration (^{134}Cs + ^{137}Cs), which was calculated on a dry weight basis. Radiocesium deposition data are from soil surveys carried out concurrently at the same plantations (Forestry Agency, 2012b).

3. Results and discussion

The radioactive contamination caused by the accident in March 2011 was so heavy that effects of atmospheric nuclear tests or Chernobyl accident were negligible in this study.

Radiocesium deposition (*i.e.*, the total activity in the organic layer and surface soil (0–5 cm)) was highly correlated with the air dose rate in 2011 (Fig. 1). The concentration of radiocesium in male flowers in 2011 was also correlated to the air dose rate, measured at 1 m above ground, and their relationship could be approximated by a logarithmic linear equation. The measured concentration ranged between < 0.1 kBq kg^{-1} and 260 kBq kg^{-1} (Fig. 2).

The male flowers were not directly exposed to the fallout from the March 2011 accident because they would have developed during July-October 2011. Thus, the radiocesium detected in the male flowers must have been translocated from other parts of the tree. Sugi is an evergreen conifer that holds its needles in the canopy for several years. Mitsui *et al.* (1955) showed that squash plants can absorb radioactive material through the leaves, so the radiocesium detected in the male sugi flowers may have been translocated from needles that had been exposed to the radioactive fallout. It is also possible that the radiocesium in the male flowers was originally absorbed by the tree roots. Although the sugi root system develops under the organic layers on forest floor, radiocesium that had passed through the organic layers into the mineral soil might have been absorbed by sugi roots near the soil surface.

In 2012, radiocesium activities of male flowers, current-year needles, one-year-old needles and old needles were also logarithmic linear to radiocesium deposition in 2011 (Fig. 3). The average radioactivity levels decreased in the order old needles, one-year-old needles, male flowers, and current-year needles. Yoshida *et al.* (2011) revealed that the youngest needles and branches of pine tree contained higher ^{137}Cs and stable Cs than older ones in Belarus in July 1998. We attribute the high radioactivity of old needles to direct exposure to the fallout; a considerable amount of the radio-

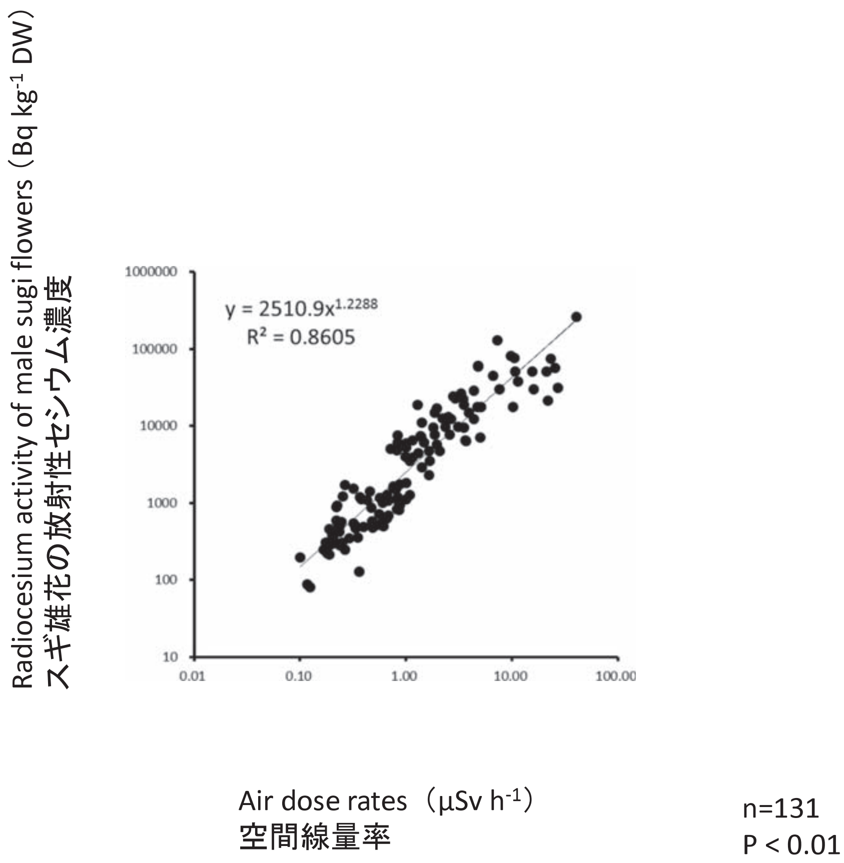

Fig. 2. Correlation between radiocesium activity of male sugi flowers and air dose rates in 2011 (air dose rates were measured at 1 m above ground level)
図–2. スギ雄花中の放射性セシウム濃度と空間線量率の関係（2011年）

cesium still remained in 2012 on the old needles. Table 1 shows correlations between radiocesium activities of male flowers, current-year needles, one-year-old needles and old needles of sugi. They have strong correlations each other.

The air dose rates on the 24 sugi plantations in 2012 were logarithmic linear to the rates on those plantations in 2011 (Fig. 4). The 2012 values were approximately 90% of the 2011 values, although the decay of radiocesium for this period predicts approximately 80%. Radiocesium migration in the forest ecosystem may explain the difference between the

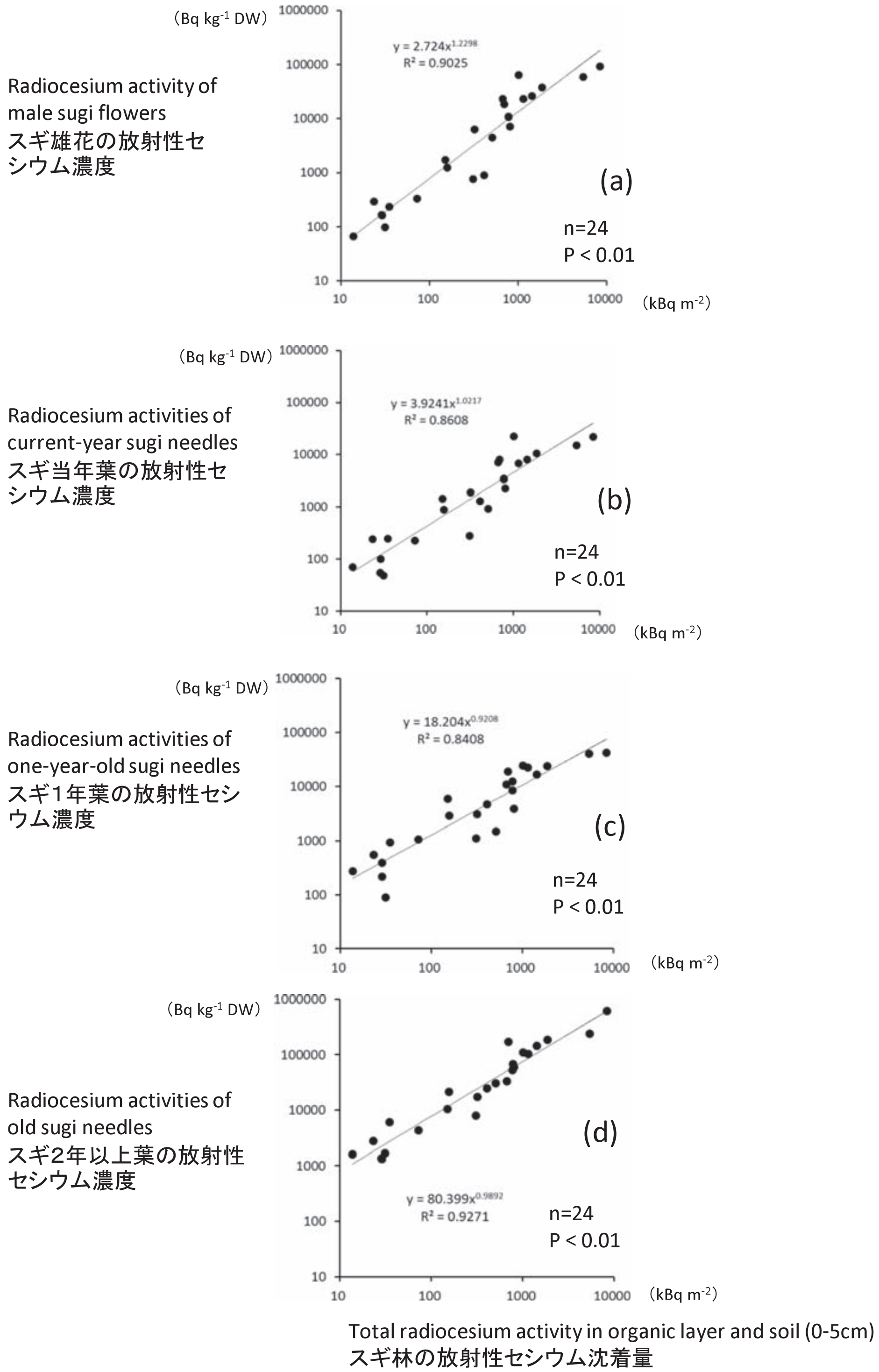

Fig. 3. Correlation between radiocesium activities of male flowers (a), current-year needles (b), one-year-old needles (c), old needles (d) of sugi in 2012 and total radiocesium deposition (total activity in the organic layer and surface soil (0−5 cm)) in 2011

図−3. スギ林の放射性セシウム沈着量(2011年の堆積有機物層と土壌(0−5cm)の合計)とスギ雄花，当年葉，1年葉，2年以上葉中の放射性セシウム濃度(2012年)との関係

Tabe 1. Correlation between radiocesium activities of male flowers, current-year needles, one-year-old needles and old needles of sugi n = 24

表−1. スギの雄花，当年葉，１年葉，２年以上葉中の放射性セシウム濃度の相互関係

Y \ X	male flowers in 2011 雄花2011	male flowers in 2012 雄花2012	current−year needles in 2012 当年葉	one−year−old needles in 2012 1年葉	old needles in 2012 2年以上葉
male flowers in 2012 雄花2012	$y = 0.4427x^{0.9901}$ $R^2 = 0.9457$		$y = 0.762x^{1.1521}$ $R^2 = 0.9604$	$y = 0.1442x^{1.2208}$ $R^2 = 0.8969$	$y = 0.0171x^{1.2051}$ $R^2 = 0.9146$
current−year needles in 2012 当年葉	$y = 0.7939x^{0.8325}$ $R^2 = 0.9238$	$y = 1.6679x^{0.8336}$ $R^2 = 0.9604$		$y = 0.2153x^{1.0708}$ $R^2 = 0.9535$	$y = 0.0433x^{1.0304}$ $R^2 = 0.9242$
one−year−old needles in 2012 1年葉	$y = 4.4927x^{0.7457}$ $R^2 = 0.8913$	$y = 9.6256x^{0.7347}$ $R^2 = 0.8969$	$y = 5.7385x^{0.8905}$ $R^2 = 0.9535$		$y = 0.3104x^{0.9296}$ $R^2 = 0.9046$
old needles in 2012 2年以上葉	$y = 21.66x^{0.7796}$ $R^2 = 0.9309$	$y = 51.691x^{0.759}$ $R^2 = 0.9146$	$y = 35.769x^{0.8969}$ $R^2 = 0.9242$	$y = 8.1375x^{0.973}$ $R^2 = 0.9046$	

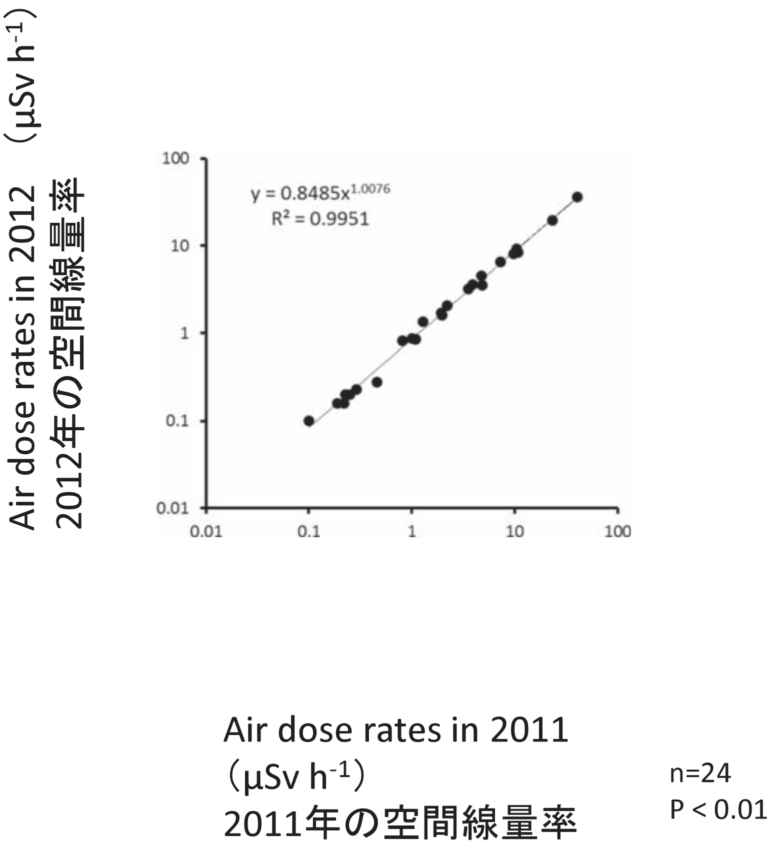

Fig. 4. Comparison of air dose rates between 2011 and 2012

図−4. 2011年から2012年への空間線量率の変化

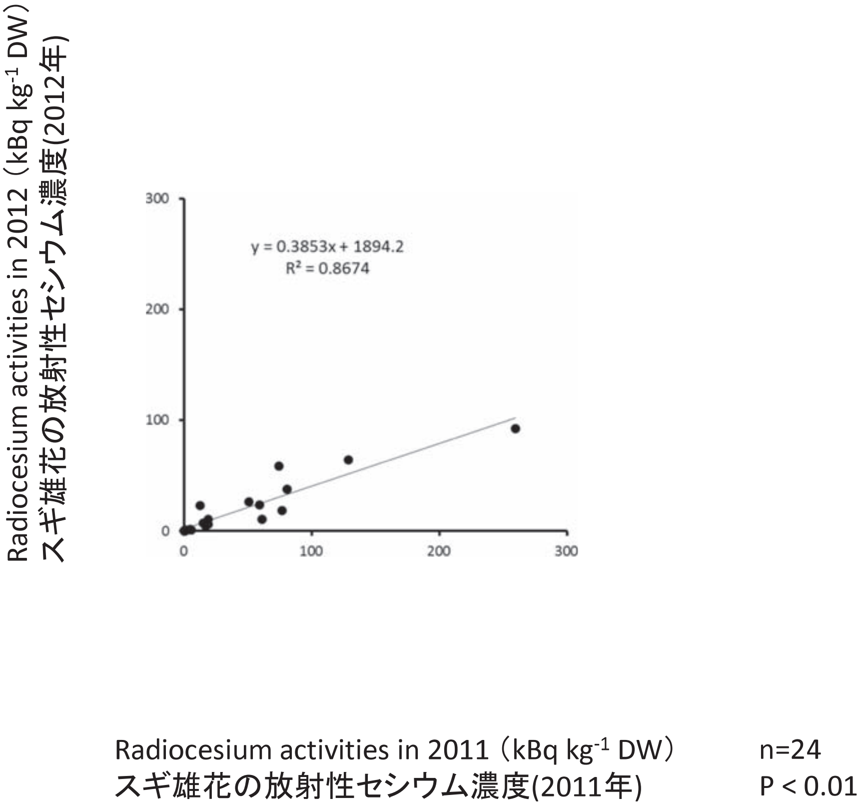

Fig. 5. Comparison of total radiocesium activities of male sugi flowers between 2011 and 2012

図−5. 2011年と2012年のスギ雄花中の放射性セシウム濃度の比較

estimates on disintegration of radionuclides and the measured air dose rates. Litter falls increase air dose rates because radiocesium on the old needles in the canopies comes close to the measuring point of air dose rates (*i.e.*, a height of one meter).

The maximum radiocesium activity in male flower in 2012 was 92.6 kBq kg^{-1}, compared with 260 kBq kg^{-1} in 2011. Radiocesium activities of male flowers measured in 2012 were proportional to those measured in 2011, but on average the 2012 values were about 40% of the 2011 values (Fig. 5). The sugi trees apparently did not absorb much radiocesium in 2012. In one male flower sample, the radiocesium activity exceptionally increased from 2011 to 2012, but the reason for this increase is not clear. The radiocesium in the soil (0−5 cm) of this plantation, however, was over ten times the amount of the radiocesium in the organic layer on the forest floor.

The radioactivities of male flowers, current-year needles and one-year-old needles showed logarithmic linear relationships with the radiocesium deposition in the soil. These relationships suggest that the absorption of radiocesium by the trees was not selective and did not depend on level of biological activity. Rather, the amount absorbed mainly reflected the radiocesium concentration in the environment. Thus, radiocesium released by the accident was apparently passively absorbed by the trees, mainly during a short time interval after the accident, and subsequent absorption was less.

The Forestry Agency (2012a) used the available data, including the air dose rates and the concentration of radiocesium in male flowers in Fig. 2, to predict sugi pollen dispersal and estimated that the spread of radiocesium from the forest via sugi pollen dispersal was probably negligible.

4. Acknowledgment

The authors thank the Fukushima Prefectural Forestry Research Centre for their cooperation. This work was supported by research grant # 201126 from FFPRI, and the Forestry Agency.

Literature cited

Forestry Agency (2012a) http://www.rinya.maff.go.jp/j/press/hozen/120208.html (Only in Japanese).

Forestry Agency (2012b) http://www.rinya.maff.go.jp/j/press/hozen/120301.html (Only in Japanese).

Goor, F. *et al.* (2007) Radiocaesium accumulation in stemwood: Integrated approach at the scale of forest stands for contaminated Scots pine in Belarus. Journal of Environmental Management 85: 129−136.

Hashimoto, S. *et al.* (2013) Predicted spatio-temporal dynamics of radiocesium deposited onto forests following the Fukushima nuclear accident. Scientific Reports (Nature publishing group) 3: 2564, DOI: 101038/srep02564.

Hirono, Y. and Nonaka, K. (2012) Translocation and distribution of radiocesium in tea plants. *In* Proceedings of International Science Symposium on Combating Radionuclide Contamination in

Agro-soil Environment: 380.

Kato, H. *et al.* (2012) Interception of the Fukushima reactor accident-derived ^{137}Cs, ^{134}Cs and ^{131}I by coniferous forest canopies. Geophysical Research Letters 39(20), DOI: 10.1029/2012GL052928.

Koarashi, J. *et al.* (2012) Factors affecting vertical distribution of Fukushima accident-derived radiocesium in soil under different land-use conditions. Science of the Total Environment 431:392–401.

Kuroda, K. *et al.* (2013) Radiocesium concentrations in the bark, sapwood and heartwood of three tree species collected at Fukushima forests half a year after the Fukushima Dai-ichi nuclear accident. Journal of Environmental Radioactivity 122: 37–42.

Matsunaga, T. *et al.* (2013) Comparison of the vertical distributions of Fukushima nuclear accident radiocesium in soil before and after the first rainy season, with physicochemical and mineralogical interpretations. Science of the Total Environment 447:301–314.

Mitsui, S. *et al.* (1955) Investigations on the radioactive contamination of crop plants as a result of H-bomb detonation (Part 2) root and foliage uptake of "Bikini ash". Soil and plant food 1:17–18.

Tagami, K. *et al.* (2012) Translocation of radiocesium from stems and leaves of plants and effect on radiocesium concentrations in newly emerged plant tissues. Journal of Environmental Radioactivity 111:65–69.

Thiry, Y. *et al.* (2009) Impact of Scots pine (*Pinus sylvestris* L.) plantings on long term ^{137}Cs and ^{90}Sr recycling from a waste burial site in the Chernobyl Red Forest. Journal of Environmental Radioactivity 100:1062–1068.

Yoshida, S. *et al.* (2011) Distribution of radiocesium and stable elements within a pine tree. Radiation Protection Dosimetry: pp. 1–4.

Yoshihara, T. *et al.* (2013) Radiocesium contaminations of 20 wood species and the corresponding gamma-ray dose rates around the canopies at 5 months after the Fukushima nuclear power plant accident. Journal of Environmental Radioactivity 115:60–68.

Zhiyanski, M. *et al.* (2010) Cesium-137 contamination of oak (*Quercus petrae* Liebl.) from sub-mediterranean zone in South Bulgaria. Journal of Environmental Radioactivity 101:864–868.