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

ARTICLE *in* JOURNAL OF ENVIRONMENTAL RADIOACTIVITY · AUGUST 2012

Impact Factor: 2.48 · DOI: 10.1016/j.jenvrad.2012.07.002 · Source: PubMed

4 AUTHORS:

Toshihiro Yoshihara
Central Research Institute of Electric Power…

Hideyuki Matsumura
Central Research Institute of Electric Power…

Shin-nosuke Hashida
Central Research Institute of Electric Power…

Toru Nagaoka
Central Research Institute of Electric Power…

Journal of Environmental Radioactivity 115 (2013) 60–68

Contents lists available at SciVerse ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

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

Toshihiro Yoshihara*, Hideyuki Matsumura, Shin-nosuke Hashida, Toru Nagaoka

Plant Molecular Biology, Laboratory of Environmental Science, Central Research Institute of Electric Power Industry (CRIEPI), 1646 Abiko, Chiba 270-1194, Japan

ARTICLE INFO

Article history:
Received 17 May 2012
Received in revised form
3 July 2012
Accepted 6 July 2012
Available online xxx

Keywords:
Woody plants
Evergreen species
Deciduous species
Radiocesium
Fukushima

ABSTRACT

Radiocesium ($^{134}Cs + ^{137}Cs$) deposition from the Fukushima nuclear power plant accident was measured in 20 woody plants (12 evergreen and 8 deciduous species) grown in Abiko (approximately 200 km SSW from the NPP). Leaves (needles) and twigs were sampled from each of three foliar positions (top, middle, and bottom) in the plant canopy in early August 2011. At the time, soils around the plants were also sampled, and gamma radiation dose rates were measured at each sampling position. The average radiocesium activity in the observed leaves of the evergreen species was 7.7 times that in the leaves of the deciduous species. Among the observed evergreen coniferous species, the activity in pre-fallout-expanded leaves was 2.4 times that in the post-fallout-expanded leaves. Notably, a distinct variation in the activity among the evergreen coniferous species could be observed for the post-fallout-expanded leaves but not for the pre-fallout-expanded leaves. Although these differences depend on whether the leaves had expanded at the time of the fallout, it is probable that a considerable amount of radiocesium was translocated to newly developed leaves at a species-specific rate. In addition, it was demonstrated that dose rates around woody plants were not consistent with the prevailing prediction that general dose rates correspondingly decrease with monitoring height from the ground. Thus, the dose rates in the top foliar layer of the deciduous species decreased more than predicted, whereas those in the top foliar layer of the coniferous species did not decrease. This may be due to differences in the balance between the attenuation resulting from a shielding effect of the plant bodies and the higher radiocesium accumulation in the leaves.

1. Introduction

A large earthquake and tsunami struck northeastern Japan on 11 March 2011. The accident of Fukushima nuclear power plant (NPP) that resulted from the earthquake and tsunami contaminated large areas with radionuclides consisting primarily of ^{131}I, ^{134}Cs, and ^{137}Cs (MEXT, 2012). Forests and woody plants were the major recipients of the terrestrial fallout. For example, some estimations from the earlier surveys indicates that forest occupies at least 1343 km^2 of the total contaminated areas of 1778 km^2 with more than 5 mSv y^{-1} (MOE, 2012) and totally 21 Tg-Dry matter of forest components are contaminated (Hashimoto et al., 2012). Additionally, based on their physiological/morphological aspects, forests/woody plants seemed to be objects that warranted particular attention because woody plants are effective aerosol interceptors (Bunzl and Kracke, 1988; Sokolov et al., 1990; Petroff et al., 2008; Pröhl, 2009). Plants growing on the forest floor have roots that are largely confined to the acidic, organic-rich humic soil layer where the bioavailability of radiocesium is relatively high (McGee et al., 2000; Goor and Thiry, 2004). In forests, the natural decontamination of radionuclides is extremely slow because of their low net export (less than 1%) to the outside environment (Tikhomirov and Scheglov, 1994; Nylén, 1996). Many areas of forests/woody plants are located close to residential areas, especially in city areas. Forests/woody plants have several important functions, such as providing water, food, a timber supply and recreation areas, indicating that it is necessary to monitor the level and migration of contamination for the utilization of their products and/or the admittance into the areas for several decades.

From the Chernobyl accident in 1986, we can learn about the distribution and fate of radionuclides in the environment. The observations of radionuclide dispersal were conducted not only for the neighboring district of Chernobyl (Tikhomirov and Scheglov,

Abbreviations: NPP, Nuclear power plant.

* Corresponding author. Tel.: +81 4 7182 1181; fax: +81 4 7183 3347.
E-mail address: yoshiha@criepi.denken.or.jp (T. Yoshihara).

0265-931X/$ – see front matter © 2012 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.jenvrad.2012.07.002

1994; Fogh and Andersson, 2001; Sokolik et al., 2001; Ramzaev et al., 2006), but also for most parts of Europe, including Belgium (Ronneau et al., 1991; Sombré et al., 1994), Bulgaria (Zhiyanski et al., 2008), England (Bonnet and Anderson, 1993), France (Barci-Funel et al., 1995), Germany (Bunzl and Kracke, 1988), Greece (Clouvas et al., 2005), Spain (Navasa et al., 2012), and Sweden (Melin et al., 1994; Fawaris and Johanson, 1995; McGee et al., 2000; Belyazida et al., 2006). These observations not only provided information regarding the regional and local characteristics of the radionuclide distribution, but they also provided information related to the science associated with forest ecosystems/woody plants, such as plant physiology, meteorology, soil science and physico-chemistry, geomorphology, and radiology. However, many differences exist between the accidents in Fukushima and in Chernobyl. First, the contaminated area and the total amount of fallout from the Fukushima accident were fortunately much smaller than those of Chernobyl (e.g., total ^{131}I and ^{137}Cs fallout to air from the Fukushima accident were 1.3–1.6 $\times$ 10^{17} Bq and 1.1–1.5 $\times$ 10^{16} Bq, respectively, whereas those from the Chernobyl accident were 1.8 $\times$ 10^{18} Bq and 8.5 $\times$ 10^{16} Bq, respectively; MEXT, 2012). Although the major long-lived radio-contaminants in the soil resulting from both the Chernobyl and Fukushima accidents are ^{137}Cs and ^{134}Cs, the Chernobyl soil is also contaminated with other nuclides, such as ^{90}Sr, $^{239+240}$Pu and ^{241}Am, at non-negligible levels (Hoshi et al., 1994; Sokolik et al., 2001). Second, the fate of radionuclides in forests is most likely influenced by the climate, soil texture, and vegetation in terms of their weathering and/or accumulation properties (Pröhl, 2009; Tagami et al., 2011). In the present study, we reported the radiocesium accumulation and the corresponding gamma radiation dose rates in/around 20 woody plant species at the end of the first growing season (early August 2011) after the Fukushima NPP accident. It could not only provide a primary status of the radiocesium contamination in some major plant species in Japan, but also show clear differences in the accumulation property among species. We lastly discussed the results in relation to the surrounding dose rates.

2. Materials and methods

2.1. The study site

Measurements of gamma radiation dose rates and samplings of plants and soils were carried out in our laboratory in Abiko, Japan (35.87815, 140.02487), which is located approximately 200 km SSW from the NPP in Fukushima (37.41868, 141.02215). Abiko has a moderate monsoon climate (average annual total rainfall of 1481.5 mm between 2001 and 2011; average annual daily average, annual minimum, and annual maximum air temperatures of 15.2, −4.0, and 35.9 °C, respectively, between 2001 and 2011. The specific changes in daily rainfall, daily averaged air temperature, and daily irradiation are provided in S Fig. 1–4, respectively). The laboratory is situated at an uptown area of flat land approximately 20 m in elevation (there is no obvious slope and no erosion/accumulation in the study site). The total area is 17.3 ha, which is close to a major river (Tone-gawa, approximately 2 km N) but is not close to the sea coast (approximately 25 km S and 60 km E) and is covered with many types of plant species, including tall woods with heights of over 20 m. Most of the target woody plants stand alone or as part of colonnades or the margins of small forests. The basal soil type is clay overlapped with a thin organic layer. Additionally, the bases of most of the woody plants were located on bare ground, although some of them were covered with grasses. Notably, the initial radionuclide fallout was observed on 16 March 2011 as dry-deposition; however, the majority of the fallout was observed on 21 March 2011 with rainfall (Morino et al., 2011).

2.2. Target woody plants

The target woody plants consisted of a total of 30 plants of 20 different species (Table 1). These are arborous or shrub species; however, Japanese wisteria (*Wisteria floribunda*, *Wf*, No. 30 in Table 1) alone belong to vines and grown as a pleach. The observed numbers, 1–3 replications per species, are not enough to see the intra-species variation; however, it is enough at least to compare the aspects between types as shown below. All of these species are very popular in Japan for landscape gardening and/or forestation. The ages of the woody plants were not specifically known; however, they were assumed to range from 10 to 30 years based on the timing of planting. The plants can be divided into four groups according to their physiological/morphological aspects, namely evergreen species (coniferous and broad leaves) and deciduous species (coniferous and broad leaves). The absolute heights of the monitoring positions are shown in Fig. 1.

Table 1
A list of the target woody plant species.

Classification	Common name (*Scientific name*), abbreviation	Total samples per species[a]
Evergreen species (coniferous)	1) **Japanese cedar** (*Cryptomeria japonica*), ***Cj1***[b]	9
	2) *ibid*., ***Cj2***	
	3) *ibid*., ***Cj3***	
	4) **Japanese red pine** (*Pinus densiflora*), ***Pd***	3
	5) **Himalayan cedar** (*Cedrus deodara*), ***Cd***	3
	6) **Hinoki cypress** (*Chamaecyparis obtusa*), ***Co***	3
	7) **Itohiba cypress** (*Chamaecyparis pisifera* var.*Filifera*), ***Cp1***	3
	8) *ibid*., ***Cp2***	
	9) *ibid*., ***Cp3***	
Evergreen species (broad leaves)	10) **Wax myrtle** (*Myrica rubra*), ***Mr***	3
	11) **Omurasaki azalea** (*Rhododendron pulchrum*), ***Rp***	1
	12) **Sasanqua** (*Camellia sasanqua*), ***Cs***	1
	13) **Camphor tree** (*Cinnamomum camphora*), ***Cc***	3
	14) **Sweet viburnum** (*Viburnum odoratissimum* var. *awabuki*), ***Vo***	3
	15) **Japanese box tree** (*Buxus microphylla var. japonica*), ***Bm***	1
	16) **Moso bamboo** (*Phyllostachys pubescens*), ***Pp***	3
Deciduous species (coniferous)	17) **Metasequoia** (*Metasequoia glyptostroboides*), ***Mg1***	6
	18) *ibid*., ***Mg2***	
Deciduous species (broad leaves)	19) **Somei-yoshino cherry tree** (*Prunus* x *yedoensis*), ***Py1***	12
	20) *ibid*., ***Py2***	
	21) *ibid*., ***Py3***	
	22) *ibid*., ***Py4***	
	23) **Horse chestnut** (*Aesculus hippocastanum*), ***Ah1***	6
	24) *ibid*., ***Ah2***	
	25) **Sycamore** (*Platanus orientalis*), ***Po***	3
	26) **American dogwood** (*Benthamidia florida*), ***Bf***	3
	27) **Crape myrtle** (*Lagerstroemia indica*), ***Li***	1
	28) **Trident maple** (*Acer buergerianum*), ***Ab1***	6
	29) *ibid*., ***Ab2***	
	30) **Japanese wisteria** (*Wisteria floribunda*), ***Wf***	1

[a] Basically, three samples grown at different foliar positions were provided per individual stand besides shrubs.
[b] Some species indicated with different numbers in the abbreviation are discriminating individuals.

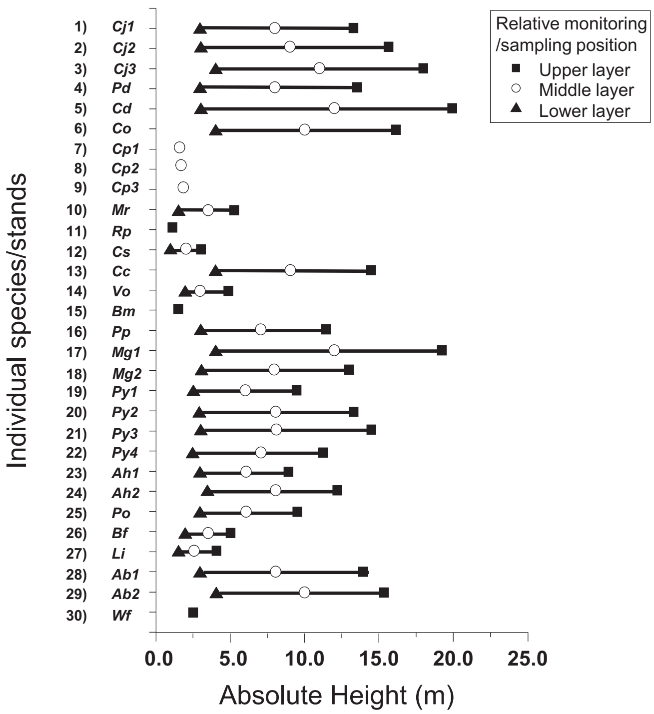

Fig. 1. Absolute height distributions for the sampling and the measurement positions absolute heights (m) from ground level of each sampling/measurement position are plotted for individual species/stands. All the name are indicated as the abbreviations (see Table 1). The relative situations of these positions are also indicated as different symbols; closed rectangle, upper foliar layer; open circle, middle foliar layer; closed triangle, lower foliar layer.

2.3. Measurement of gamma radiation dose rates, sampling, and nuclide analysis

The measurement of gamma radiation dose rates and sampling were conducted on 6–7 August 2011. The measurement of dose rate around the canopy and the sampling were carried out using a vehicle designed for working at large heights. Gamma radiation dose rates were measured using a CsI (TI) scintillation spectrometer (PDS-100GN/ID, MIRION technologies, San Ramon, CA; Dose rate accuracy is ±30% [^{137}Cs]) without collimation. At least three measurements (the spectrometer was left for 30 s, and measurements were then taken three times every 30 s, thus requiring a total of 2 min for each point) were made at each sampling point, and the average values were computed. The measurement points were the same as the sampling points shown below; they were 50 cm apart in the canopy at each sampling point or 1 m above the ground under the woody plants.

Foliar parts and twigs were sampled from each of three relative foliar positions (top, middle, and bottom) in the canopy (Fig. 1). Thus, the absolute sampling heights were different for each plants even though the relative positions were the same. The absolute height of each measurement/sampling position was detected with a laser range finder (Laser 550A, Nikon, Tokyo, Japan). Several pieces of foliar parts (leaves or needles) and twigs, corresponding to a maximum total volume of approximately 1 kg-FW, were collected at each position. At the same time, soils around the woody plants were also sampled. After the sampling, foliar samples of evergreen coniferous woody plants were separated into two groups according to their expanding season (pre-fallout-expanded leaves and post-fallout-expanded leaves) to determine the specificity in the radionuclide accumulation, while the samples of other species (post-fallout-expanded leaves or unknown leaves) and the twig samples (pre-fallout-expanded twigs) were directly used for the nuclide analysis. Every plant sample was dried for at least 3 days in an oven at 65 °C. Then, the samples were homogenized, weighed, and packed in U8 containers for the radioactivity measurements. No replicate plant samples were prepared (one sample per sampling position). On the other hand, the sampling of soils was based on a core sampling procedure using a commercial sampler (100 ml, φ 50 × 51 (L) mm). Four core soil samples located 1 m apart from each other along a circle around each woody plant were acquired as the replicates. These were packed directly from the sampler into U8 containers and weighed for the nuclide analysis. Following the analysis, samples were dried, sieved, and re-weighed to evaluate the dry matter content. A Ge-semiconductor detector (GEM20-70, ORTEC, Oak Ridge, TN; The detection limit at 3600 s observation is about 1 Bq kg-DW^{-1}) coupled with a mulch channel analyzer (MCA7600, SEIKO EG&G, Tokyo, Japan) was used to evaluate ^{134}Cs and ^{137}Cs activities in the woody plant and soil samples (MEXT, 1976). The measurement time was 3600 s unless noted otherwise. The radioactivity was determined after adjusting for the time of sampling (7 August 2011, approximately 160 days after the assumed deposition), although the actual measurement of the radioactivity ended in late January 2012.

2.4. Statistical analysis

Basically, the data showed lognormal distribution. Thus, we analyzed the data by parametric tests, that is, Tukey–Kramer's multiple comparison and Student's t-test after log-conversion. The tests were conducted using a software packages, Kyplot (ver. 4.0; KyensLab Inc., Tokyo, Japan) and Origin (ver. 6.1J; OriginLab Co., MA, USA). Differences were basically considered statistically significant at $p \leq 0.05$.

3. Results

3.1. Activity of radionuclides in woody plants

The average radiocesium (^{134}Cs and ^{137}Cs) activity among all of the observed foliar parts was 4000 Bq kg-DW^{-1}, with a range between 180 Bq kg-DW^{-1} (horse chestnut, *Ah*) and 25,000 Bq kg-DW^{-1} (Japanese box woody plant, *Bm*; Fig. 2). The average activity in twigs was 2800 Bq kg-DW^{-1}, with a range between 104 Bq kg-DW^{-1} (sycamore, *Po*) and 7400 Bq kg-DW^{-1} (American dogwood, *Bf*; Fig. 3). The specific radionuclide ratio (^{134}Cs/^{137}Cs) was approximately 0.9 at the time of sampling for each sample. In general, the activities of the foliar parts were significantly higher ($p = 0.016$; Fig. 2) in evergreen species (6100 Bq kg-DW^{-1} on average) than in deciduous species (790 Bq kg-DW^{-1} on average). However, even among evergreen species, some of the species, such as wax myrtle (*Mr*) and camphor woody plant (*Cc*), exhibited lower radiocesium activities in the foliar parts comparable to the deciduous species. On the other hand, no significant difference was observed with regard to the activities of the twigs between evergreen species (3200 Bq kg-DW^{-1} on average) and deciduous species (2400 Bq kg-DW^{-1} on average) ($p = 0.64$; Fig. 3). In addition, there was no significant difference in the radiocesium activity between all observed foliar parts and twigs when the activities were averaged for all species ($p = 0.42$).

The radiocesium activities in the foliar parts of the evergreen coniferous species were 2.4 times higher ($p \leq 0.01$) in the pre-fallout-expanded parts (pre-leaves; 6600 Bq kg-DW^{-1} on average) than in the post-fallout-expanded parts (post-leaves; 2800 Bq kg-DW^{-1} on average; Fig. 4A). Additionally, the average activity in the post-leaves of all observed evergreen coniferous species was 4.5 times higher ($p \leq 0.01$) than that in the post-leaves of all observed deciduous species (620 Bq kg-DW^{-1} on average; Fig. 4A). The total radiocesium activities in the pre-leaves ranged between 5900 (Japanese cedar, *Cj*) and 8200 Bq kg-DW^{-1} (hinoki cypress, *Co*;

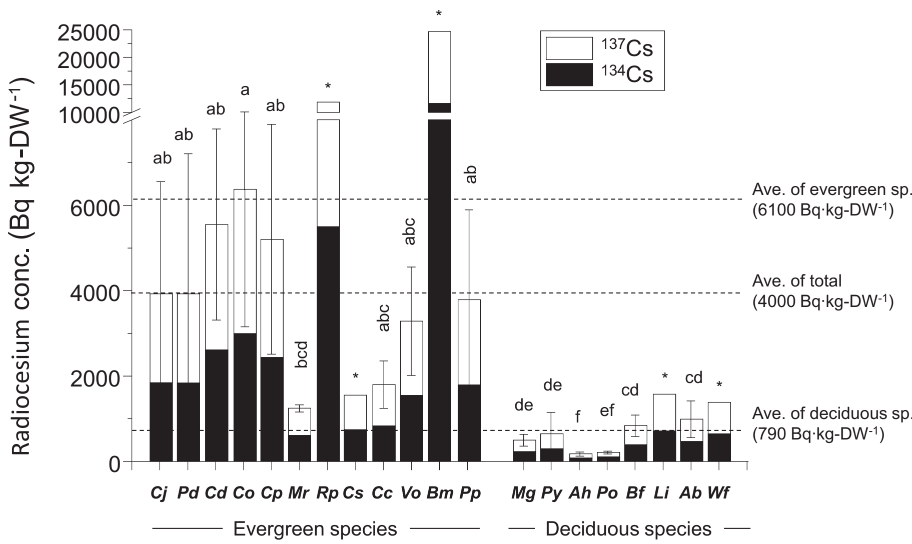

Fig. 2. Radiocesium activities in all observed foliar parts averages and standard deviations of radiocesium activities in all observed foliar parts (i.e. pre- and post-fallout-expanded leaves sampled at every position) are calculated for individual species. Data are separately indicated for the two major phenotypes, evergreen species and deciduous species. Although the averages are separately plotted for ^{134}Cs (closed rectangles) and ^{137}Cs (open rectangles), the standard deviations are indicated for their total. Different alphabets on the standard deviations demonstrate significant differences ($p \leq 0.05$) among all species for each total radiocesium activity by Tukey–Kramer's multiple comparison after log conversion. Data with asterisk instead of alphabet are excluded from the multiple comparison because of no replication.

Fig. 4B), whereas those in the post-leaves ranged between 1100 (Japanese red pine, *Pd*) and 4700 Bq kg-DW^{-1} (Himalayan cedar, *Cd*). Notably, no obvious variation among the species could be observed in the activities of the pre-leaves, although a clear variation could be observed in the activities of the post-leaves (Fig. 4B). The variation in the individual activities due to the different sampling positions was indicated as the post-/pre-leaves radiocesium activity ratio (Fig. 5). This ratio ranged between 0.14 and 0.85; however, no significant tendency was identified other than the species-dependent specificity. Furthermore, no obvious relationship was observed between the absolute sampling height and the radiocesium activity (Fig. 6).

3.2. Activity of radionuclides in soils under woody plants

The total radiocesium (^{134}Cs and ^{137}Cs) activities in the soils under the woody plants ranged between 80 (Metasequoia, *Mg1*) and 3900 Bq kg-DW^{-1} (itohiba cypress, *Cp3*) (Fig. 7). In general, the activities were higher ($p = 0.02$) in the soils under the evergreen species (1900 Bq kg-DW^{-1} on average) than in those under the deciduous species (1100 Bq kg-DW^{-1} on average); however, no particular species exhibited significant differences in the soil radiocesium activity. On the other hand, the soil radiocesium activities showed weak but significant linear correlation efficiencies with the average radiocesium activities in all observed foliar parts ($r = 0.43$, $p = 0.02$; data not shown).

3.3. Gamma radiation dose rate around woody plants

The average gamma radiation dose rate measured around the woody plant targets was 0.34 μSv hr^{-1}, with a range between 0.20 (upper position of the wax myrtle, *Mr*) and 0.49 μSv hr^{-1} (middle position of the itohiba cypress, *Cp2*, and under the Somei-yoshino cherry woody plant, *Py4*) (Fig. 8). The dose rates decreased with

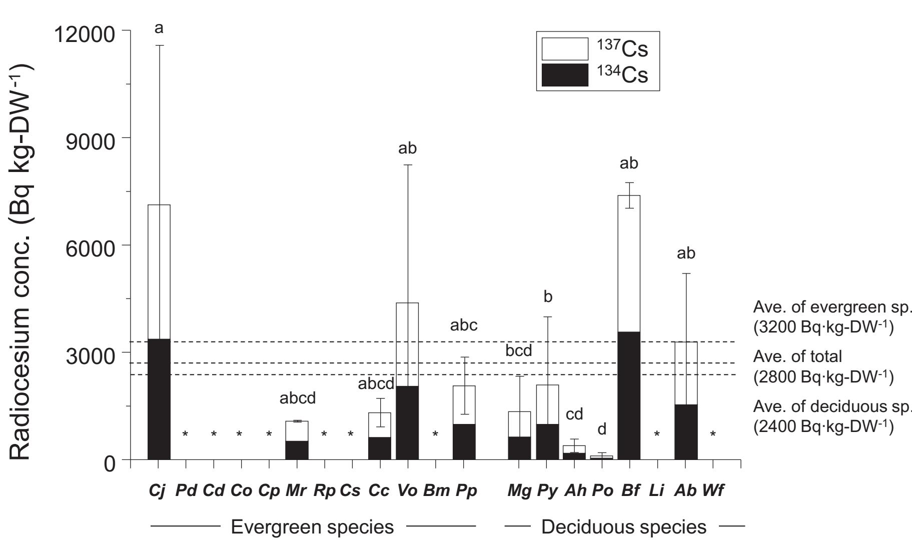

Fig. 3. Radiocesium activities in pre-fallout-expanded twigs averages and standard deviations of radiocesium activities in pre-fallout-expanded twigs are indicated. Different alphabets on the standard deviations demonstrate significant differences ($p \leq 0.05$) among all species for each total radiocesium activity by Tukey–Kramer's multiple comparison after log conversion. Data with asterisk instead of alphabet are excluded from the multiple comparison because of not-determined.

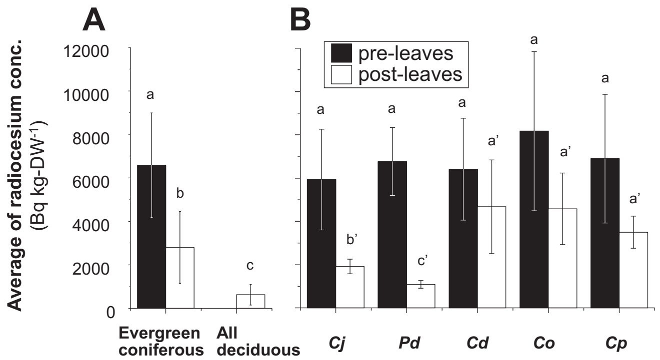

Fig. 4. Radiocesium activities in pre- and post-fallout-expanded foliar parts of evergreen coniferous species averages and standard deviations of radiocesium activities in pre- (closed rectangles) and post-fallout-expanded leaves (open rectangles) are shown for all evergreen coniferous species and all deciduous species (A), and individual evergreen coniferous species (B). Different alphabets on the standard deviations demonstrate significant differences ($p \leq 0.05$) among data by Tukey–Kramer's multiple comparison after log conversion. The calculation is separately conducted for data in (A), (B)-pre-fallout-expanded leaves, and (B)-post-fallout-expanded leaves, respectively.

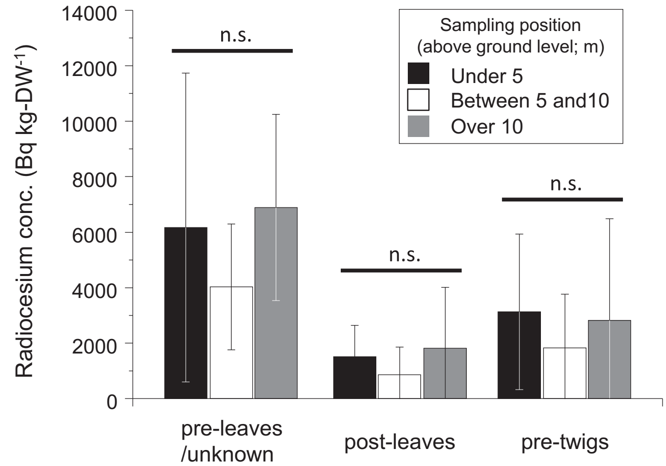

Fig. 6. Height independent distribution of the radiocesium activities radiocesium activities in pre-leaves or unknown-leaves (in cases for evergreen coniferous species and evergreen broad leaf species), post-leaves (in cases for evergreen coniferous species and deciduous species), and pre-twigs (in cases for all) are separately averaged for three different sampling positions (under 5 m, between 5 and 10 m, and over 10 m, respectively) and shown with standard deviations. No significant differences (n.s., $p \geq 0.05$) could be seen among three sampling positions by Tukey–Kramer's multiple comparison after log conversion in every sample type (pre-leaves or unknown-leaves, post-leaves, and pre-twigs).

increasing absolute height of the measurement positions. This tendency became clearer when the data were separately plotted for evergreen coniferous species and deciduous species after conversion of the absolute dose rate into the relative dose rate (dose rate at 1 m above the ground for individual plants was indicated as 1.0; Fig. 9). Although the data should follow an exponential function on a larger scale, the data were analyzed as a linear function in this report because the data were packed within a narrow range and exhibited a better fit with a linear function. The results provided a clear validation of the separation of these data into two groups from a statistical viewpoint; that is, the correlation coefficient (r) between the dose rates and the measurement height was -0.79 ($p \leq 0.01$) for deciduous species, whereas that for evergreen coniferous species was -0.32 ($p = 0.13$). In other words, the dose rates of all of the evergreen coniferous woody plants were in the same range, irrespective of the measurement height, and were therefore obviously different from those of the deciduous species.

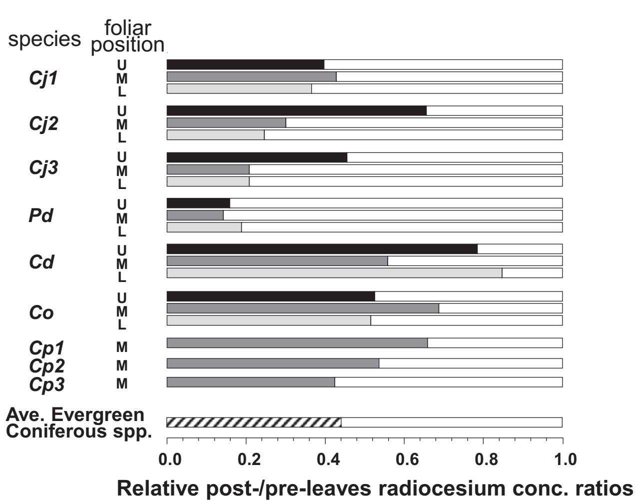

Fig. 5. Post-/pre-leaves radiocesium activity ratios relative ratios of radiocesium activity in post-leaves against pre-leaves (post-/pre-leaves ratio; each of radiocesium activity in pre-leaves is shown as 1.0) are demonstrated for each foliar position of observed evergreen coniferous species. The average of total evergreen coniferous species irrespective of the foliar position is also shown in the lowest column.

4. Discussion

4.1. Differences in radiocesium accumulation between evergreen and deciduous species

It was obvious that the radiocesium ($^{134}Cs + ^{137}Cs$) activities of the foliar parts were higher in evergreen species than in deciduous species (Fig. 2). This is because the foliar parts of evergreen species had expanded at the time of fallout but those of the deciduous species had not. This presumption is supported by the fact that no species-dependent differences could be observed in the radiocesium activities of the observed twigs, all of which were pre-fallout-expanded (Fig. 3). Such a difference in radiocesium accumulation between evergreen and deciduous species was also observed in Fukushima and another hot spot (Chiba), which is located a similar distance away from Fukushima as Abiko, although their absolute contamination levels were different (Amano et al., 2011; Forestry Agency, 2011; Tagami et al., 2011). In Fukushima, the largest radiocesium distribution in the forests of evergreen species is in the foliar layer (29–56% of the total deposition), whereas that in the forests of deciduous species is in the litter layer (66% of the total deposition). In Chiba, woody plants with old leaves that had fallen out before the fallout show higher activities than those with newly expand leaves after the fallout. On the other hand, a considerable level of radiocesium was also detected in leaves of deciduous species, which were post-fallout-expanded (post-leaves; Fig. 2). The average radiocesium level in the deciduous foliar parts reached more than one-eighth of that in the total evergreen foliar parts, meaning that such amounts of accumulation could be observed as a result of translocation from the contaminated parts (e.g., twigs, branch, and trunk) and/or the soils. Notably, when the activities in the post-leaves were compared among all observed species, they were still lower in the deciduous species than in the evergreen coniferous species (Fig. 4A). It is possible that this difference is due to the differences in the total deposition on each plant body, as well as to the differences in the translocation ability among plants. Similar evidence of radiocesium translocation was

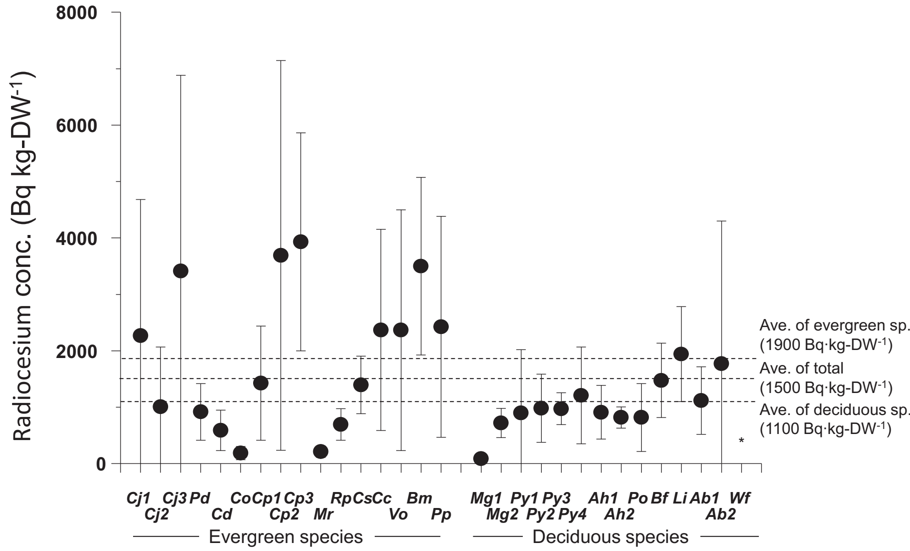

Fig. 7. Radiocesium activities in soils under the woody plants averages and standard deviations of radiocesium activities in soils under the observed woody plants are shown for individual species/stands. Data are separately indicated for the two major phenotypes, evergreen species and deciduous species. The difference between averages of the deciduous species and the evergreen species is significant ($p \leq 0.05$, by Student's *t*-test), although the difference among individual species is not (by Tukey–Kramer's multiple comparison). Asterisk indicates not-determined.

reported for the Chernobyl accident by Ronneau et al. (1991) and for the Fukushima accident by Tagami et al. (2011).

4.2. Specific radiocesium distribution among evergreen species

A considerable difference in radiocesium activity could be observed even among the foliar parts of the evergreen species (Fig. 2). For example, the wax myrtle (*Mr*) and camphor woody plant (*Cc*) exhibited significantly lower radiocesium activities, which were comparable to those found in deciduous species, than Himalayan cedar (*Cd*), hinoki cypress (*Co*), and itohiba cypress (*Cp*). When the foliar parts of evergreen coniferous species were divided into pre- and post-leaves, the relative radiocesium activity ratios (post-/pre-leaves ratios) varied among species, which could be due primarily to the variation in the activity of post-leaves (Figs. 4B and 5). These findings could not be explained on the basis of the timing of foliar expansion, as shown above, but could rather be explained on the basis of the leaf longevity and the defoliation timing. Evergreen coniferous species usually

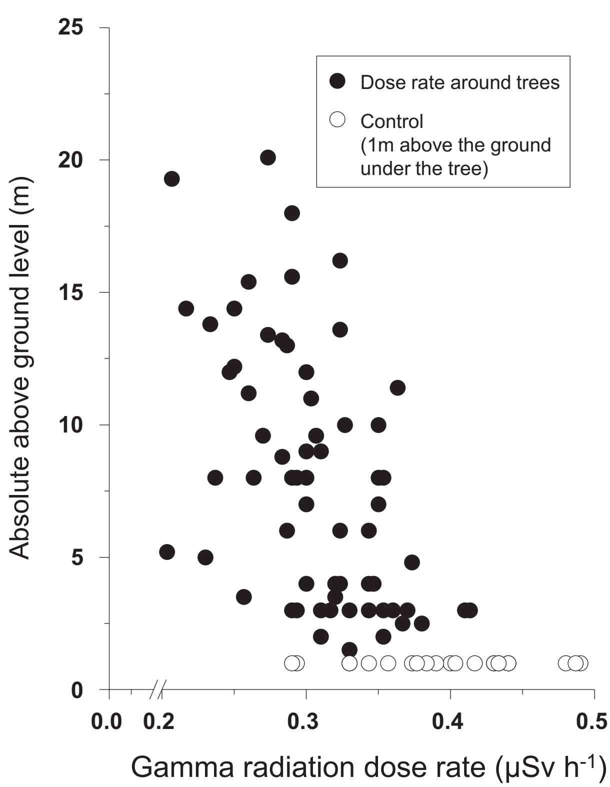

Fig. 8. Gamma radiation dose rate around the woody plants gamma radiation dose rates around the observed woody plants are plotted by a factor of absolute monitoring height (m; closed circles). The dose rates monitored 1 m above the ground under the tree are also plotted as the control (open circles).

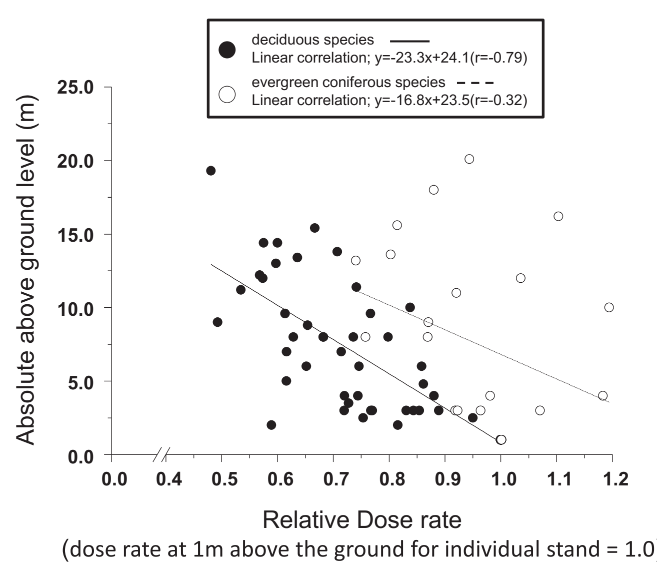

Fig. 9. Comparison of relative dose rates around the woody plants between the deciduous species and the evergreen coniferous species relative dose rates (dose rates at 1 m above the ground for individual plants are shown as 1.0) of the deciduous species (closed circle) and the evergreen coniferous species (open circle) are plotted by a factor of absolute monitoring height (*m*). The data are statistically analyzed the linearity using poison's test.

expand new leaves every spring, grow them for several years, and fall their leaves almost in concert at a season in the order of their expansion (Hagihara et al., 1978; Saito, 1990; Mediavilla and Escudero, 2003; Yamashita et al., 2004). Thus, it is probable that the observed evergreen coniferous species had already grown the majority of their pre-leaves and expanded the post-leaves at the time of sampling. In other words, the difference in the numbers of pre-/post-leaves could cause the variation in the average radiocesium activity in all observed foliar parts among the species. In contrast, ordinary evergreen broad-leaf species also expand new leaves every spring; however, the leaf longevity of these species is comparatively shorter than that of evergreen coniferous species (i.e., at most 1–2 years), and the defoliation is always gradually occurring throughout their life span, even though some of them fall their leaves in concert at a season (Escudero et al., 1992; Mediavilla and Escudero, 2003). Although the exact time at which the observed evergreen broad-leaf species fell the pre-leaves was not clear, it was possible that some of the species exhibiting lower radiocesium activities had already fallen the majority of their pre-leaves before the time of sampling. In addition, it was possible that the leaf longevity and the defoliation timing influence the radiocesium translocation from old tissues to newly developed tissues. Sombré et al. (1994) demonstrate that spruce and oak trees translocate radiocesium during the growing season from the reservoir (e.g., trunk and old foliage) to newly developed tissues, resulting in an imbalance in the activity between old and new foliage, whereas no difference in the foliage activity can be observed in winter, irrespective of their age. Sombré et al. (1994) also suggest that the translocation of cesium is a circumstantial result of the seasonal translocation of essential elements, especially potassium. Furthermore, as hypothesized by Tagami et al. (2011), another possibility is that the morphological characteristics of leaves, such as the surface and topological aspects, contributed to the activity difference through radiocesium deposition and/or weathering. Accordingly, Nitta and Ohsawa (1998) demonstrate that the winter buds of woody plants can be categorized into the following three types based on the external morphology and developmental processes: naked, hypsophyllary and scaled buds. It is possible that such a morphological characteristic, even prior to the leaf expansion, contributes to the differences in foliar radiocesium activities among species.

4.3. Radiocesium accumulation in soils under the woody plants

The radiocesium accumulation in soils was significantly higher in the soils under the evergreen species than in the soils under the deciduous species (Fig. 7). This can also be considered as an attribute of the timing of foliar expansion, as discussed above, which is reasonable based on the fact that trees are effective aerosol interceptors and that larger canopies should be able to collect lager amounts of fallout (Bunzl and Kracke, 1988; Sokolov et al., 1990; Petroff et al., 2008). Pröhl (2009) demonstrated that, due to the dependence on plant development, the interception of both dry and wet deposits is subject to pronounced seasonality. The radioactivity retained on vegetation is lost due to the weathering, which is influenced by various factors in addition to the stage of development of the plant, such as elemental properties of the radionuclide and the amount and intensity of rain during a rainfall event. Notably, when the present data were compared to data taken at a neighboring farm (MEXT, 2012), the former (1500 kBq kg^{-1} on average) showed higher radiocesium activity than the latter (220 kBq kg^{-1} on average). The tendency seemed suitable to our claim. However, the difference may mainly come from the sampling soil depth (the present data showed averages of 5 cm soil depth, whereas the MEXT data shows that of 30 cm soil depth). We could not ruled out a possibility that the sampling scheme adopted or biased by the difference in stemflow/throughfall. In relation to this consideration, a precise observation of the forest contamination in Fukushima indicates that, although the contamination ranged between 0.7 and 4580 kBq kg^{-1}, the variation is independent of the dominant species in the forest (Forestry Agency, 2011). Thus, the radiocesium activity in the fallout would be a more important factor of the contamination than the dominant species in the forest. Additionally, the fact that a larger amount of the radiocesium in the evergreen forest in Fukushima is still located in the foliar layer than in the forest floor (Forestry Agency, 2011) may be related to the different distribution tendencies identified in the present study. It is probable that such tendencies are not always applicable to all cases; that is, the physico-chemical properties of the soil, such as the soil consistency/permeability, the texture of the soil (which consists of minerals and organic matter), and the status of the coverage (i.e., litter and understory), may also be an important factor associated with the radiocesium accumulation (Rühm et al., 1996; Sokolik et al., 2001).

4.4. Correlation between radiocesium accumulation and gamma radiation dose rate

The gamma radiation dose rate around the observed woody plants substantially decreased with the monitoring height from the ground (Fig. 8). This tendency itself has previously been predicted (Eckerman and Ryman, 1993; IAEA, 2000; Iwamoto et al., 2011). In brief, the absorption of unscattered radiation by air attenuates the dose rate in proportion to the distance from the source; however, scattered radiation conversely increases the dose rate when the contamination is dispersed over a substantially broad and deep soil area. In this case, the decrease in the dose rate is far more gradual than in the case of a point source contamination. Based on the assumption that the contaminated ground is shaped in a circle with a 400-m radius and that the soil within a 1-m depth is uniformly contaminated with ^{137}Cs at 1000 kBq m^{-2}, Iwamoto et al. (2011) predicted that the attenuation of the dose rate is one-half to one-third for every 50 m increase in height from the ground. On the other hand, the results of the present study were not consistent with the prevailing prediction with regard to two of the findings. The first inconsistency is that a larger decrease in the dose rates around deciduous species was observed than predicted (Fig. 9). The dose rate around deciduous species decreased to approximately half even when the monitoring height was 10 m above the ground level. This could be due to a considerable attenuation of the dose rate by a shielding effect of the plant bodies in the case of the deciduous species. Another inconsistency is that no decrease in the dose rates around evergreen coniferous species was observed in the present study. This could be due to a higher radiocesium accumulation in the leaves of the evergreen coniferous species.

No relationship could be found between the absolute sampling heights and the radiocesium activities (Fig. 6). However, Fogh and Andersson (2001) demonstrate that a maximum radiocesium activity in tree is found in a specific height of pine trunk and suggest that the vertical transport of cesium inside the stem is of a magnitude that is insufficient to eliminate height differences in activity. In the present observation, a few evergreen coniferous woody plants (i.e., Japanese cedar, *Cj*) demonstrated higher radiocesium activities in leaves sampled at higher positions than in those sampled at lower positions (data not shown). Although most of the tree parts uniformly intercepted the radiocesium, the possibility that some of the parts accumulate more radiocesium than other parts cannot be ruled out.

5. Conclusions

Radiocesium accumulation was measured in 20 woody plant species in Abiko, which is located approximately 200 km SSW from the NPP, to determine the resting amounts of the radionuclide at the end of the first growing season after the NPP accident in Fukushima. The primary findings of the study are as follows: 1. The radiocesium activities of the foliar parts were higher in evergreen species than in deciduous species, which is due to the fact that the foliar parts of the evergreen species had expanded at the time of fallout but those of the deciduous species had not. Additionally, it is probable that the leaf longevity and the defoliation timing were associated with the differences among the evergreen species, although the possibility that the morphological characteristics of the trees contributed to the differences cannot be ruled out. 2. Radiocesium accumulation was also detected in the post-fallout-expanded tissues, irrespective of the species, at various activities. This suggested a species-specific translocation of radiocesium in their bodies or uptake from the soils. 3. The radiocesium accumulation in soils was significantly higher under the evergreen species than under the deciduous species. Even though the tendency would be variable by case (i.e., various factors, such as pant developmental stages, elemental properties and amounts of the radionuclide, intensity of rain, and possibly a sampling scheme may affect the tendency), the present case can also be considered as an attribute of the timing of foliar expansion, as discussed above. 4. The dose rates in the top foliar layer of deciduous species decreased more than predicted, whereas those in the top foliar layer of coniferous species did not decrease. This may be due to differences in the balance between the attenuation resulting from a shielding effect of the plant bodies and the higher radiocesium accumulation in the leaves. These findings could be useful for a prospective forest/garden management/activity to avoid the detrimental effects of radioactivity on our health, such as safety forest products certification and proper radionuclides elimination program. Further and continuous investigations are necessary to determine how long and how much radiocesium accumulates in the canopy and under the woody plants.

Acknowledgments

We thank Mr. Hiroshi Shimura, Ms. Miki Ueda and Ms. Mari Sato (Co. Ltd. Ceres), Mr. Yukihiro Taemi and Mr. Keita Yamaguchi (Co. Ltd. Denryoku Tech. Sys.) for their skillful assistance in the measurement of gamma radiation dose rate, sampling, and radionuclides analysis.

Appendix A. Supplementary material

Supplementary data related to this article can be found online at http://dx.doi.org/10.1016/j.jenvrad.2012.07.002.

References

Amano, H., Akiyama, M., Chunlei, B., Kawamura, T., Kishimoto, T., Kuroda, T., Muroi, T., Odaira, T., Ohta, Y., Takeda, K., Watanabe, Y., Morimoto, T., 2011. Radiation measurements in the Chiba Metropolitan area and radiological aspects of fallout from the Fukushima Dai-ichi nuclear power plants accident. J. Environ. Radioact.. Published ahead of print.

Barci-Funel, G., Dalmasso, J., Barci, V.L., Ardisson, G., 1995. Study of the transfer of radionuclides in trees at a forest site. Sci. Total Environ. 173/174, 369–373.

Belyazida, S., Westlingb, O., Sverdrup, H., 2006. Modelling changes in forest soil chemistry at 16 Swedish coniferous forest sites following deposition reduction. Environ. Pollut. 144, 596–609.

Bonnet, P.J.P., Anderson, M.A., 1993. Radiocaesium dynamics in a coniferous forest canopy: a mid-Wales case study. Sci. Total Environ. 136, 259–277.

Bunzl, K., Kracke, W., 1988. Cumulative deposition of ^{137}Cs, $^{238, 239, 240}$Pu and ^{241}Am from global fallout in soils from forests, grassland and arable land in Bavaria (FRG). J. Environ. Radioact. 8, 1–14.

Clouvas, A., Xanthos, S., Antonopoulos-Domis, M., Alifragis, D.A., 2005. Radiocesium gamma dose rates in a Greek pine forest: measurements and Monte Carlo computations. Radioact. Environ. 7, 1155–1166.

Eckerman, K.F., Ryman, J.C., 1993. External Exposure To Radionuclides in Air, Water, and Soil, Federal Guidance Report No. 12, EPA-402-R-93-081, p. 234.

Escudero, A., del Arco, J.M., Sanz, I.C., Ayala, J., 1992. Effects of leaf longevity and retranslocation efficiency on the retention time of nutrients in the leaf biomass of different woody species. Oecologia 90, 80–87.

Fawaris, B.H., Johanson, K.J., 1995. A Comparative study on radiocaesium (^{137}Cs) uptake from coniferous forest soil. J. Environ. Radioact. 28, 313–326.

Fogh, C.L., Andersson, K.G., 2001. Dynamic behavior of ^{137}Cs contamination in trees of the Briansk region, Russia. Sci. Total Environ. 269, 105–115.

Forestry Agency, 2011. A Press Release on 27 Dec. 2011: Radionuclides Distribution in Forests (No. 2). http://www.rinya.maff.go.jp/j/press/hozen/pdf/111227_2-01.pdf (Final access 03.05.12).

Goor, F., Thiry, Y., 2004. Processes, dynamics and modelling of radiocaesium cycling in a chronosequence of Chernobyl-contaminated Scots pine (*Pinus sylvestris* L.) plantations. Sci. Total Environ. 325, 163–180.

Hagihara, A., Suzuki, M., Hozumi, K., 1978. Seasonal fluctuations of litter fall in a *Chamaecyparis obtuse* plantation. J. Jpn. Forestry Soc. 60, 397–404.

Hashimoto, S., Ugawa, S., Nanko, K., Shichi, K., 2012. The total amounts of radioactivity contaminated materials in forests in Fukushima, Japan. Sci. Rep. 2, 416. http://dx.doi.org/10.1038/srep00416.

Hoshi, M., Yamamoto, M., Kawamura, H., Shinohara, K., Shibata, Y., Kozlenko, M.T., Takatsuji, T., Yamashita, S., Namba, H., Yokoyama, N., 1994. Fallout radioactivity in soil and food samples in the Ukraine: measurements of iodine, plutonium, cesium, and strontium isotopes. Health Phys. 67, 187–191.

IAEA, 2000. Generic Procedures for Assessment and Response during a Radiological Emergency, IAEA-TECDOC-1162, pp. 1–186.

Iwamoto, Y., Satoh, D., Endo, A., Sakamoto, Y., Kureta, M., Kugo, T., 2011. Study on Soil Decontamination and Dose Rate Reduction Effect, JAEA-Technology 2011-026, pp. 1–18.

McGee, E.J., Synnott, H.J., Johanson, K.J., Fawaris, B.H., Nielsen, S.P., Horrill, A.D., Kennedy, V.H., Barbayiannis, N., Veresoglou, D.S., Dawson, D.E., Colgan, P.A., McGarry, A.T., 2000. Chernobyl fallout in a Swedish spruce forest ecosystem. J. Environ. Radioact. 48, 59–78.

Mediavilla, S., Escudero, A., 2003. Leaf life span differs from retention time of biomass and nutrients in the crowns of evergreen species. Funct. Ecol. 17, 541–548.

Melin, J., Wallberg, L., Suomela, J., 1994. Distribution and retention of cesium and strontium in Swedish boreal forest ecosystems. Sci. Total Environ. 157, 93–105.

MEXT (Ministry of Education, Culture, Sport, Science, and technology), 1976. The manual for analysis of radiocesium. In: Issue No. 3 of a Series of Manuals for Measurement of Radioactivity. Japan chemical analysis center, Chiba, Japan, pp. 21–27.

MEXT (Ministry of Education, Culture, Sport, Science, and technology), 2012. Monitoring Information Of Environmental Radioactivity Level. http://radioactivity.mext.go.jp/en/ (Final access 03.05.12).

MOE (Ministry of Environment), 2012. The 2nd Skull Session for Environmental Recovery from the Great East Japan Earthquake. www.env.go.jp/jishin/rmp/conf/02-mat4.pdf (Final access 20.02.12).

Morino, Y., Ohara, T., Nishizawa, M., 2011. Atmospheric behavior, deposition, and budget of radioactive materials from the Fukushima Daiichi nuclear power plant in March 2011. Geophys. Res. Lett. 38, L00G11. http://dx.doi.org/10.1029/2011GL048689.

Navasa, A., López-Vicentea, M., Gaspar, L., Machín, J., 2012. Assessing soil redistribution in a complex karst catchment using fallout ^{137}Cs and GIS. Geomorphology. published ahead of print.

Nitta, I., Ohsawa, M., 1998. Bud structure and shoot architecture of canopy and understorey evergreen broad-leaved woody plants at their Northern limit in east Asia. Ann. Bot. 81, 115–129.

Nylén, T., 1996. Uptake, turnover and transport of radiocaesium in boreal forest ecosystems, thesis Swedish Univ. Agr. Sci. Uppsala Sweden, ISBN: 91-576-5149-3.

Petroff, A., Mailliat, A., Amielh, M., Anselmet, F., 2008. Aerosol dry deposition on vegetative canopies. part I: review of present knowledge. Atmos. Environ. 42, 3625–3653.

Pröhl, G., 2009. Interception of dry and wet deposited radionuclides by vegetation. J. Environ. Radioact. 100, 675–682.

Ramzaev, V., Andersson, K.G., Barkovsky, A., Fogh, C.L., Mishine, A., Roed, J., 2006. Long-term stability of decontamination effect in recreational areas near the town Novozybkov, Bryansk Region, Russia. J. Environ. Radioact. 85, 280–298.

Ronneau, C., Sombre, L., Myttenaere, C., Andre, P., Vanhouche, M., Cara, J., 1991. Radiocaesium and potassium behaviour in forest woody plants. J. Environ. Radioact. 14, 259–268.

Rühm, W., Kammerer, L., Hiersche, L., Wirth, E., 1996. Migration of ^{137}Cs and ^{134}Cs in different forest soil layers. J. Environ. Radioact. 33, 63–75.

Saito, H., 1990. Seasonal leaf-fall trends in Japanese red pine stands and their synchronization among stands. Sci. Rep. Kyoto Pref. Univ. For. 42, 47–57.

Sokolik, G.A., Ivanova, T.G., Leinova, S.L., Ovsiannikova, S.V., Kimlenko, I.M., 2001. Migration ability of radionuclides in soil-vegetation cover of Belarus after Chernobyl accident. Environ. International 26, 183–187.

Sokolov, V.E., Rjabov, I.N., Tikhomirov, F.A., Shevchenko, V.A., Taskaev, A.I., 1990. Ecological and Genetic Consequences of the Chernobyl Atomic Power Plant accident. ICSU, SCOPE-RADPATH, Lancaster Radpath Meeting, University of Lancaster, UK, 26–30 March 1991.

Sombré, L., Vanhouche, M., de Brouwer, S., Ronneau, C., Lambotte, J.M., 1994. Long-term radiocesium behaviour in spruce and oak forests. Sci. Total Environ. 157, 59–71.

Tagami, K., Uchida, S., Ishii, N., Kagiya, S., 2011. Translocation of radiocesium from stems and leaves of plants and the effect on radiocesium activities in newly expanded plant tissues. J. Environ. Radioact.. Published ahead of print.

Tikhomirov, F.A., Scheglov, A.I., 1994. Main investigation results on the forest radioecology in the Kyshtym and Chernobyl accident zones. Sci. Total Environ. 157, 45–57.

Yamashita, T., Kasuya, N., Nishimura, S., Takeda, H., 2004. Comparison of two coniferous plantations in central Japan with respect to forest productivity, growth phenology and soil nitrogen dynamics. Forest Ecol. Manage. 200, 215–226.

Zhiyanski, M., Bech, J., Sokolovska, M., Lucot, E., Bech, J., Badot, P.-M., 2008. Cs-137 distribution in forest floor and surface soil layers from two mountainous regions in Bulgaria. J. Geochem. Explor. 96, 256–266.