Interception of the Fukushima reactor accident-derived137Cs, 134Cs and 131I by coniferous forest canopies

Interception of the Fukushima reactor accident-derived ^{137}Cs, ^{134}Cs and ^{131}I by coniferous forest canopies

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[1] The Fukushima Daiichi nuclear power plant accident resulted in extensive radioactive contamination of the surrounding forests. In this study, we analyzed fallout ^{137}Cs, ^{134}Cs, and ^{131}I in rainwater, throughfall, and stemflow in coniferous forest plantations immediately after the accident. We show selective fractionation of the deposited radionuclides by the forest canopy and contrasting transfer of radiocesium and ^{131}I from the canopy to the forest floor in association with precipitation. More than 60% of the total deposited radiocesium remained in the canopy after 5 month of the initial fallout, while marked penetration of the initially deposited ^{131}I through the canopy was observed. The half-lives of ^{137}Cs absorbed in the cypress and cedar canopies were calculated as 620 days and 890 days, respectively for the period of 0–160 days. The transfer of the deposited radiocesium from the canopy to the forest floor was slow compared with that of the spruce forest affected by fallout from the Chernobyl nuclear reactor accident. **Citation:** Kato, H., Y. Onda, and T. Gomi (2012), Interception of the Fukushima reactor accident-derived ^{137}Cs, ^{134}Cs and ^{131}I by coniferous forest canopies, *Geophys. Res. Lett.*, *39*, L20403, doi:10.1029/2012GL052928.

1. Introduction

[2] The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident occurred on March 12 and 14, 2011, after the Great East Japan Earthquake on March 11, 2011. The accident released high concentrations of nuclides into the atmosphere [*Butler*, 2011; *Chino et al.*, 2011]. Gamma-ray-emitting radionuclides such as cesium-137 (^{137}Cs), cesium-134 (^{134}Cs), and iodine-131 (^{131}I) were diffused in the atmosphere [*Amano et al.*, 2012; *Hirose*, 2012] and widely deposited on ground surfaces throughout northeast Japan (http://radioactivity.mext.go.jp/en/).

[3] Radionuclides were highly dispersed in surrounding forested areas, as 70% of the land area in Japan is covered by forest; this is also true of Fukushima and the neighboring prefectures. Most of the forested areas are covered by evergreen conifers such as Japanese cypress (*Chamaecyparis obtusa*) and cedar (*Cryptomeria japonica*) plantations [*Onda*

et al., 2010]. Thus, to elucidate the behavior of the deposited radionuclides in such forest plantationsis of key importance.

[4] In the vegetated areas, a fraction of the radionuclides deposited on the vegetation was initially intercepted and retained by the canopy [*Hoffman et al.*, 1995; *Pröhl and Hoffman*, 1996; *Kinnersley et al.*, 1996, 1997] and then transferred to the ground surface as a result of weathering processes [*Bunzl et al.*, 1989; *Bonnett and Anderson*, 1993; *Pröhl*, 2009]. In a spruce forest in Munich, Germany, which was affected by the fallout of the Chernobyl accident-derived radiocesium, the half-life of ^{134}Cs absorbed in the canopy was calculated as 90 days for the period 0–130 days after the initial atmospheric deposition [*Bunzl et al.*, 1989]. In contrast, the behavior of ^{131}I in aquatic environments depends on its chemical form [*Muramatsu et al.*, 1987; *Uchida et al.*, 1989]. However, the interception and retention characteristics of the deposited radionuclides by different tree species, especially radioiodine, immediately after the accidental fallout have not been understood.

[5] When and how the radionuclides on the forest canopy were mobilized to the ground surface remains unknown. The process and mechanisms of distribution and transfer of deposited radionuclides are difficult to determine during the crisis; however, these have critical implications for radiation hygiene, decontamination and modeling biogeochemical cycling. In this study, we report the initial behavior of the ^{137}Cs, ^{134}Cs and ^{131}I deposited in cypress and cedar forests immediately after the accidental release of radioactive materials from the FDNPP.
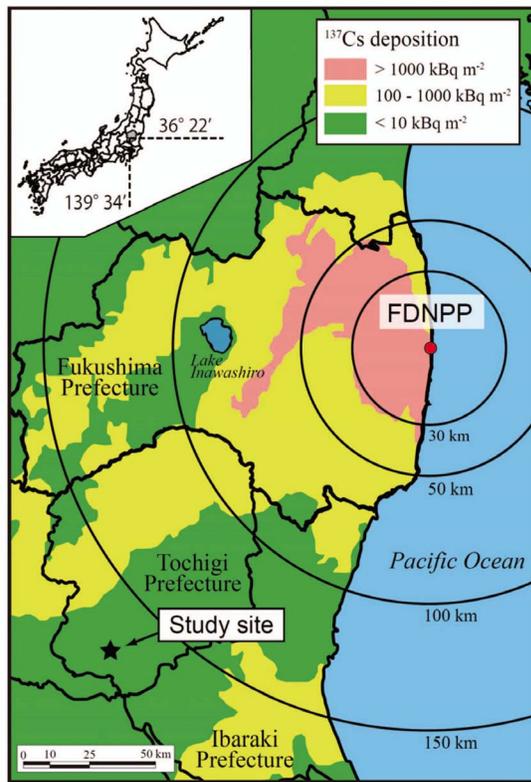
2. Materials and Methods

[6] The study site was located in Tochigi Prefecture 150 km southwest of the FDNPP (Figure 1). The total deposition of FDNPP-derived ^{137}Cs at the study site was estimated to be less than 10,000 Bq m^{-2} based on the results of an airborne monitoring survey by MEXT (http://radioactivity.mext.go.jp/en/). The annual precipitation in this area is 1270 mm, and the mean annual temperature is 14.2°C (data obtained from 2001 to 2010 at Yamakoshi, the nearest national weather station). Experimental plots were selected in Japanese cypress (*Chamaecyparis obtusa*) and Japanese cedar (*Cryptomeria japonica*) forests in the study site. The stand density of the 40-year-old cypress forest was 2500/ha, whereas that of the 41-year-old cedar forest was 1300/ha. The slope gradient for both plots was 30 degrees.

[7] Twenty throughfall collectors in a lattice-like pattern were located in the experimental plot. Each throughfall collector consisted of a 2-L bottle and a 13-cm-diameter funnel. The throughfall collector was equipped with an evaporation suppressor and shading film to prevent the

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(^{137}Cs) were measured using a high-purity n-type germanium coaxial gamma-ray detector (EGC25-195-R, Canberra-Eurisys, Meriden, Connecticut, USA) coupled to an amplifier (PSC822, Canberra-Eurisys) and a multichannel analyzer (DSA1000, Canberra). The measurement system was calibrated using standard gamma sources with different sample heights and was certified by the IAEA worldwide open proficiency test for the determination of gamma-emitting radionuclides (IAEA-CU-2006). All measurements of radioactivities were corrected for the radioactive decay between sample collection and radiation detection.

3. Results and Discussion

3.1. Rainfall, Throughfall, and Stemflow During the Observation Period

[9] The sampling period for each hydrological compartment was from March 11 to August 19, 2011, and included 12 rainfall events ranging from 5.3 to 184 mm of total precipitation (Table 1). The throughfall rate (total throughfall divided by total rainfall) in the cypress forest ranged from 0.50 to 0.81, with a mean of 0.71, whereas that in the cedar forest ranged from 0.37 to 0.85, with a mean of 0.75. The stemflow rates (total stemflow divided by total rainfall) for the cypress and cedar forests ranged from 0.04 to 0.14 and from 0.00 to 0.04, respectively. Therefore, 81.1% of the rainfall in the cypress forest and 77.0% of that in the cedar forest reached the ground surface via either throughfall or stemflow.

3.2. Deposition of Radionuclides by Rainfall, Throughfall, and Stemflow

[10] The atmospheric deposition of ^{137}Cs, ^{134}Cs, and ^{131}I during the observation period was 8030 Bq m^{-2}, 7510 Bq m^{-2}, and 29200 Bq m^{-2}, respectively. The ranges of ^{137}Cs, ^{134}Cs, and ^{131}I deposition during each observation period were 0.0–5420 Bq m^{-2}, 0.0–5150 Bq m^{-2}, and 820–28400 Bq m^{-2}, respectively (Table 1). Of the total local fallout during the observation period, 68% of ^{137}Cs and 97% of ^{131}I fallout occurred as rainfall during the first sampling period March 11–28. The deposition of radiocesium in rainfall decreased exponentially over time, whereas that of ^{131}I became undetectable after the second sampling period because of its short half-life (8 days).

Figure 1. Location of the study site and position of the observation instruments in the experimental plots.

evaporation of water before sampling. Rainwater was also sampled at an open site near the experimental plot to represent the atmospheric deposition flux of radionuclides at the study site. Total rainwater in the collectors was measured using a graduated cylinder after every rainfall event and sampled in a polyethylene bottle. The bottles and funnels of the collectors were washed with distilled water and wiped after sampling.

[8] The radioactivity in the water samples was measured using gamma-ray spectroscopy. Gamma-ray emissions at energies of 604 keV (^{134}Cs), 636 keV (^{131}I), and 662 keV

Table 1. Amount of Rainfall, Throughfall, Stemflow, and Radionuclides Fallout at Forest Floor During Each Sampling Period[a]

| Sampling Period | Total Amount (mm) | | | | | | | ^{137}Cs Deposition (Bq m^{-2}) | | | | | ^{134}Cs Deposition (Bq m^{-2}) | | | | | ^{131}I Deposition (Bq m^{-2}) | | | | |
| | Cypress | | | | Cedar | | | | Cypress | | Cedar | | | Cypress | | Cedar | | | Cypress | | Cedar | |
	RF	TF	SF	TF/RF	TF	SF	TF/RF	RF	TF	SF	TF	SF	RF	TF	SF	TF	SF	RF	TF	SF	TF	SF
2011/3/11–3/28	21.2	15.3	2.85	0.72	13.9	0.56	0.66	5420	434	18.5	389	12.5	5150	385	24.0	354	14.0	28400	19400	2470	13800	527
2011/3/28–4/1	9.24	7.42	0.73	0.80	5.67	-	0.61	1010	389	9.47	266	-	1010	327	5.34	231	-	821	n.d.	n.d.	n.d.	-
2011/4/1–4/13	5.27	2.62	0.21	0.50	1.93	-	0.37	400	180	36.0	90.1	-	359	189	30.9	76.8	-	n.d.	n.d.	n.d.	n.d.	-
2011/4/13–4/27	24.9	14.9	2.29	0.60	12.7	0.10	0.51	328	265	6.64	280	0.24	328	242	7.27	262	0.49	n.d.	n.d.	n.d.	n.d.	n.d.
2011/4/27–5/20	54.2	34.5	7.38	0.64	36.1	0.99	0.67	345	483	n.d.	374	10.0	277	405	n.d.	370	4.68	n.d.	n.d.	n.d.	n.d.	n.d.
2011/5/20–5/28	43.7	29.6	5.92	0.68	29.7	0.52	0.68	111	200	7.88	146	2.32	121	175	5.93	117	2.42	n.d.	n.d.	n.d.	n.d.	n.d.
2011/5/28–5/30	64.6	44.0	7.65	0.68	45.5	1.28	0.70	71.2	137	n.d.	114	n.d.	n.d.	141	21.2	112	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2011/5/30–6/13	31.3	22.3	2.66	0.71	20.1	0.10	0.64	270	234	n.d.	329	0.85	195	216	n.d.	295	0.80	n.d.	n.d.	n.d.	n.d.	n.d.
2011/6/13–6/22	41.7	29.1	5.11	0.70	31.6	0.44	0.76	75.6	112	n.d.	202	1.89	68.6	92.2	n.d.	156	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2011/6/22–7/18	79.5	52.4	8.26	0.66	53.1	0.84	0.67	n.d.	328	40.7	414	n.d.	n.d.	270	n.d.	357	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2011/7/18–7/22	184	150	15.4	0.81	157	4.96	0.85	n.d.	52.9	n.d.	141	n.d.	n.d.	13.2	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
2011/7/22–8/19	159	112	11.2	0.70	131	5.68	0.83	n.d.	96.7	n.d.	32.5	n.d.	n.d.	n.d.	n.d.	47.1	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Total	719	514	70	0.71	539	15	0.75	8030	2910	119	2780	28	7510	2460	95	2380	22	29200	19400	2470	13800	527

[a]The values in the table were Rainfall (RF), the mean of 20 measuring points for throughfall (TF), and the mean of 5 samplers for stemflow (SF); n.d. represents the deposition amount was below detection level.

[11] Coniferous forests are known to be very efficient as filters of airborne ^{131}I and radiocesium under condition of dry deposition [e.g., *Bunzl et al.*, 1989; *Livens et al.*, 1992]. *Bunzl et al.* [1989] observed 20% higher radiocesium deposition in a spruce forest compared to that in a nearby grassland. On the other hand, *Livens et al.* [1992] reported that the ^{131}I present in the soil and vegetation samples exceeds that in precipitation by about a factor of 3. However, for atmospheric deposition after the Fukushima reactor accident, many researchers estimated that most of the Fukushima derived radionuclides which were transported from the Fukushima Daiichi NPP by northeast wind encountered a band of rain that caused wet deposition in the Kanto district during March 15–16 and 21–23, 2011 [e.g., *Hirose*, 2012; *Amano et al.*, 2012; *Katata et al.*, 2012; *Srinivas et al.*, 2012; *Terada et al.*, 2012]. For ^{131}I, efficient washout from the atmosphere by precipitation was revealed for both the Chernobyl and Fukushima fallout by previous studies [e.g., *Aoyama et al.*, 1986, 1987; *Ronneau et al.*, 1967; *Landis et al.*, 2012; *Yang and Guo*, 2012]. Thus, we assumed that radiocesium and radioiodine were removed from the atmosphere through a similar pathway and wet precipitation was mainly responsible for their removal.

[12] The total deposition densities of ^{137}Cs, [134]Cs, and ^{131}I by throughfall during the observation period was 2910, 2460, and 19400 Bq m^{-2} for the cypress forest, whereas the respective values for the cedar forest were 2780, 2380, and 13800 Bq m^{-2}. By contrast, the total ^{137}Cs, [134]Cs, and ^{131}I via stemflow was 119, 95, and 2470 Bq m^{-2} for the cypress forest and 28, 22, and 527 Bq m^{-2} for the cedar forest, respectively. Although the total deposition densities of radiocesium by throughfall varied for each sampling period, throughfall showed the greatest total deposition densities of radiocesium until the end of April. The radiocesium content in stemflow was 1–2 order of magnitude smaller than that in throughfall and was often undetectable. Although ^{131}I was detected from all the throughfall and stemflow samples collected on March 28, whereas it was not detected after March 28 because of its short half-life (8 days). There was an interval of 1 month (2 months at maximum) between sample collection and radionuclide analysis, therefore a delayed measurement can cause reduction of ^{131}I concentration below the detection level even if ^{131}I exists at the time of sampling. The percentage of ^{131}I content in throughfall during the first sampling period was 68% for the cypress and 49% for the cedar forest.

3.3. Interception and Retention of Deposited Radionuclides by the Forest Canopy

[13] Our detailed monitoring revealed that radionuclides dispersed atmospherically after the accident behave differently depending on the individual radionuclide species. The total deposition densities of ^{137}Cs, [134]Cs and ^{131}I via rainfall, throughfall, and stemflow were compared to assess the interception fraction of the atmospherically deposited radionuclides by the forest canopy. During the first rainfall event between March 11 and March 28, the amount of radiocesium that reached the forest floor did not exceed 10% of the total deposited. In contrast, 75% and 49% of ^{131}I reached the forest floor by a combination of throughfall and stemflow in the cypress and cedar stand, respectively. At the end of the observation period, a simple budget calculation of the deposited radionuclides revealed that 62.3 ± 15.8%

(cypress) and 65.0 ± 18.2% (cedar) of total deposited radiocesium remained in the canopy 160 days after the accident (Figure 2). By contrast, the interception of ^{131}I differed between the tree species (25.1 ± 2.7% for the cypress and 50.9 ± 7.4% for the cedar). Nevertheless, interception of ^{131}I by the forest canopy was much lower than that of radiocesium.

[14] The differing interception rates of ^{131}I and radiocesium can be attributed to differences in the chemical properties of radionuclides. Plant surfaces are negatively charged [*Keppel*, 1966; *Ertel et al.*, 1992], therefore, anionic substances are not readily retained by the plant surface and are rapidly removed from leaf surfaces during rainfall [e.g., *Hoffman et al.*, 1995]. Similarly, the observed data in this study indicated that cationic radiocesium has a higher affinity to the forest canopy, whereas the deposited ^{131}I moved through the canopy via hydrological pathways with rainwater.

[15] For the first sampling period of March 11–28, the deposition of ^{131}I onto the forest floor for the cypress stand (68.3 ± 7.3%) was similar to the mean throughfall rate for the corresponding time period (72.1%). Conversely, for the cedar stand, the percentage of ^{131}I deposition (47 ± 7.1%) was much lower than the throughfall rate (65.6%). Although the throughfall rates were similar, differences of up to 20% for ^{131}I deposition onto the cypress and cedar forest floors were observed. The reason for this remains unknown.

[16] It is possible that the deposited ^{131}I was not readily retained by the plant surface and was being removed during runoff [*Hoffman et al.*, 1995], supporting the results observed for the cypress stand in this study. The adsorption characteristic of iodine to plant surfaces is associated with the iodine form. Organic iodine, such as CH_3I, has low solubility in water and tends to penetrate plant bodies [*Muramatsu et al.*, 1987]. Therefore, the similarity in the interception ratios for deposited ^{131}I and rainwater indicates that the deposited ^{131}I was in soluble inorganic forms. It was suggested that data on the initial interception of the deposited ^{131}I can be used to estimate its chemical forms at the time of deposition.

[17] Nevertheless, a higher interception rate of wet deposited ^{131}I for the cedar forest suggests that the potential for ^{131}I uptake by leaves varies between the cypress and cedar forest. Indeed it was reported by *Schynowski and Schwack* [1996] that the iodine concentration of the leaf cuticle of plants varies significantly with plant species. On the other hand, *Hoffman et al.* [1995] showed that the wash-off of wet deposited ^{131}I on leaves is substantially reduced if a period of drying occurs between subsequent rainfall events. Therefore, further analysis is required to clarify the influence of chemical form of the deposited ^{131}I and precipitation patterns during the first observation period, and tree species on the chemical reactions and diffusions into leaves and barks.

3.4. Dynamics of Deposited Radiocesium in a Coniferous Forest

[18] After May, total deposition of radiocesium onto the forest floor via throughfall and stemflow exceeded atmospheric flux by rainfall. This suggests that leaching of radiocesium from the canopy (rather than accumulation) occurred in the studied forests. This delayed deposition of radionuclides under forest canopies with respect to atmospheric
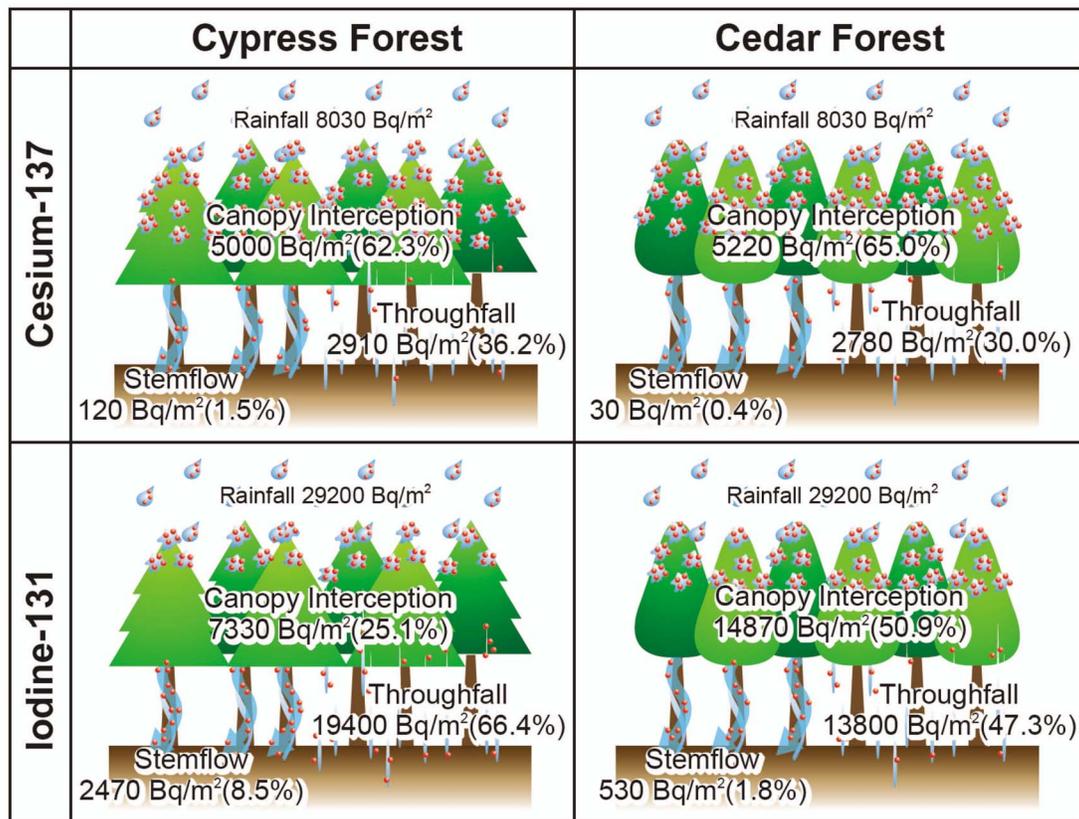
Figure 2. Schematic diagram of ^{137}Cs, ^{134}Cs, and ^{131}I deposition in rainwater, throughfall, and stemflow in the cypress and cedar forests.

input has been reported in a Norway spruce forest [*Bunzl et al.*, 1989]. The Norway spruce stand, which was 85 years old and had a stand density of 622/ha, was located in a flat area 40 km northwest of Munich, Germany, where mean annual precipitation was 800 mm. Total local ^{137}Cs deposition after the Chernobyl reactor accident was 20 kBq m^{-2}. The initial interception rate for ^{137}Cs was 70% for atmospheric deposition during April 27–30, 1986.

[19] The ^{137}Cs deposition at the forest floor occurring by throughfall and stemflow was plotted against time after the accidental release of radionuclides (Figure 3). Although the level of radioactive contamination, tree species, and climatic conditions differed between the cypress and cedar stands in this study and those in the Norway spruce stand, temporal changes in cumulative ^{137}Cs deposition at the forest floor exhibited logarithmic increases in all cases. These data indicate that in addition to the atmospheric input of radionuclides, the radionuclides in the canopy were effectively removed and transferred to forest floor over time.

[20] The half-life of ^{137}Cs absorbed in the canopy during the observation period (160 days after the beginning of radioactive fallout) was calculated as 620 days for the cypress and 890 days for the cedar stand, whereas the corresponding value for the Norway spruce was roughly 100 days. The longer half-life of ^{137}Cs in the cypress and cedar stands indicated that the removal of radionuclides through absorption by the cypress and cedar stands was slower than that by the Norway spruce stand.

[21] The total amount of radiocesium initially intercepted by the forest canopy was calculated as 5.0 kBq m^{-2} for the

cypress stand (92% of total radiocesium fallout), 5.2 kBq m^{-2} for the cedar stand (93% of total fallout), and 14 kBq m^{-2} for the Norway spruce stand (70% of total fallout). Some fractions of retained radiocesium were readily removed from the plant surface in association with rainfall; however, the remaining radiocesium was strongly absorbed by leaf, branch, and bark surfaces and was not leached from the plant surface by rainwater [*Rauret et al.*, 1994]. Nevertheless, leaf fall due to tree phenology and physical
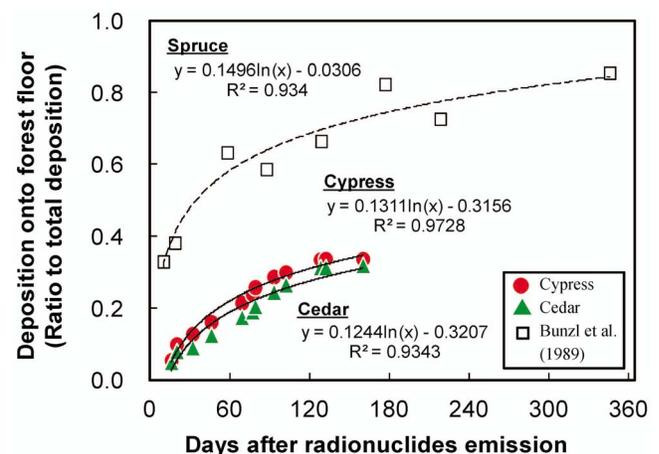
Figure 3. Deposition of radiocesium over time in coniferous forest canopies.

breakdown by heavy storms may accelerate radiocesium transfer from the canopy to forest floor [e.g., *Fesenko et al.*, 2001].

4. Conclusion

[22] This study showed the selective fractionation of the deposited radionuclides by a forest canopy and resulting contrastive transfer of radiocesium and ^{131}I from the canopy to forest floor via throughfall and stemflow. For the radiocesium, more than 60% of the total deposition remained in the canopy 5 months after the accident, while the deposited ^{131}I moved through the canopy via hydrological pathways with rainwater. Although similar interception of radiocesium was observed for the cypress and cedar forests, the interception of ^{131}I differed between the tree species (25.1% for the cypress and 50.9% for the cedar). It was suggested that data on the initial interception of the deposited ^{131}I can be used to estimate its chemical forms at the time of deposition, however further analysis is required to clarify the influence of chemical form of the deposited ^{131}I and tree species on the potential for radionuclide uptake by leaves. In addition to that, dry deposition on the forest and direct absorption of gaseous and particulate radionuclides by natural surfaces such as leaves, stems, and barks should be determined for the precise determination of interception fraction of the deposited radionuclides by the forest canopy.

[23] The high interception fraction of the deposited radiocesium by coniferous canopies indicated that the canopy will act as a secondary source of radioactive contamination of the forest floor. Furthermore, burning of the contaminated wood (e.g., forest fire) can be a source of air contamination by ^{137}Cs and ^{134}Cs. Thinning of a forest can be one of the possible ways to reduce contamination of the forest ecosystems, however a governmental decontamination policy for forests excluding areas near residential zones has not been decided because of low priority of the forest decontamination, lack of storage spaces for the radioactive waste, and limited information on distribution and transfer of the deposited radionuclides in a forest environment. The information in this paper is useful for direct measurement and radioecological modeling understanding the behavior of radiocesium and ^{131}I deposited over forest canopies in the present and future. This will provide critical information relative to the policy decision making and implementation of effective decontamination measures in the contaminated forests.

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