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ARTICLE

Temporal changes in the transfer of accidentally released ^{137}Cs from tree crowns to the forest floor after the Fukushima Daiichi Nuclear Power Plant accident

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Forests in Fukushima and neighboring prefectures accumulated atmospheric fallout of radioactive materials after the Fukushima Daiichi Nuclear Power Plant accident. We investigated the initial behavior of ^{137}Cs deposited in coniferous forest plantations. Furthermore, we conducted in-situ measurements of radioactivity in coniferous and broadleaf forest canopies. The results of this study demonstrated that more than 60% of the Fukushima reactor accident–derived radiocesium remained in coniferous forest canopies 5 months after initial deposition. Furthermore, in-situ measurements of radiocesium indicated that the radiocesium absorbed in forest canopies has been moving downward through the canopy to the forest floor.

Keywords: Fukushima Daiichi Nuclear Power Plant accident; forest canopy; interception; transfer; ^{137}Cs

1. Introduction

Radioactive materials released accidentally from the Fukushima Daiichi Nuclear Power Plant (FDNPP) were widely deposited on the land surface throughout northeastern Japan [1,2]. In Fukushima and neighboring prefectures, approximately 70% of the land area is covered by forest. The forest consists of large areas of coniferous plantations, including Japanese cedar and Japanese cypress [3], and secondary mixed forests with broadleaved and pine trees. Understanding the behavior of radionuclides deposited in forest environments is thus important.

Radioactive contamination of the forest environment was a significant environmental concern after the Chernobyl reactor accident [4]. Bunzl et al. [5] reported on the initial behavior of radionuclides deposited in an old spruce forest in Munich, which was affected by radioactive fallout from the Chernobyl reactor accident. The canopy initially intercepted and retained radionuclides deposited on the forest; subsequent transfer of radionuclides from the canopy to the forest floor occurred as a result of weathering processes (rain and wind). Canopy interception of deposited radionuclides is a very common process on vegetated surfaces [e.g., 6–10]. However, when and how radionuclides in the forest canopy are moved to the ground surface in the affected area remains a very important concern following the Fukushima reactor accident.

In this study, we investigated the behavior of radiocesium (^{137}Cs) deposited in forest environments immediately after the Fukushima reactor accident. Concentrations of ^{137}Cs in rainwater, throughfall, and stemflow were monitored in Tochigi Prefecture to determine canopy interception of atmospherically deposited radionuclides and their subsequent transfer from the canopy to the forest floor. Furthermore, at the Fukushima experimental site, which accumulated relatively greater atmospheric deposition of ^{137}Cs (300–600 kBq m^{-2}), the vertical distributions of aerial dose and gamma ray count rates derived from ^{137}Cs were measured by a portable germanium gamma-ray detector at different heights in the canopy between July 2011 and May 2012.

2. Materials and methods

2.1. Study area

We selected study sites in two areas with different contamination levels due to fallout from the Fukushima reactor accident. One site was located in Sano City in Tochigi Prefecture, 150 km southwest of the FDNPP, and the other site was located in Yamakiya District in Kawamata Town, Fukushima Prefecture, about 40 km northwest of the FDNPP (**Figure 1**). The total atmospheric deposition of ^{137}Cs after the Fukushima reactor accident was less than 10 kBq m^{-2} at the Tochigi site, whereas it was 300–600 kBq m^{-2} at the Fukushima site, based on the results of an airborne monitoring survey of radioactive contamination [2].

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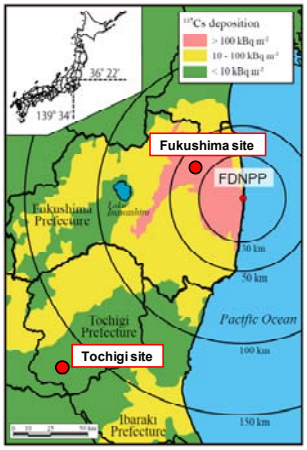

Figure 1. Location of the study site.

2.2. Experimental design

2.2.1 Tochigi site

At the Tochigi site, Japanese cypress and Japanese cedar stands were selected for study. Stand densities of Japanese cypress and Japanese cedar were 2500/ha and 1300/ha, respectively. The mean diameters of trees at breast height were 22.7 cm in the cypress stand and 26.6 cm in the cedar stand. The experimental plots are both located on hillslopes with a slope gradient of 30 degrees.

Hydrological components such as open rainfall, throughfall, and stemflow were collected during each rainfall event. An open rainfall collector equipped with an evaporation suppressor was installed at an open site near the experimental plots. Twenty throughfall collectors with evaporation suppressors were distributed in a lattice-like structure within a 10 × 10-m experimental plot, and stemflow collectors were attached to five trees in the plot. The monitoring period was 11 March to 19 August 2011. The ^{137}Cs concentration in each water sample was determined by gamma spectrometry. All radioactivity measurements were corrected for the time of sample collection.

2.2.2 Fukushima site

At the Fukushima site, young and mature cedar stands and mixed broadleaf forests of beech and red pine were selected for study. Densities of the young and mature cedar stands were 3300/ha and 1200/ha, respectively. The broadleaf forest consisted of a mixture of Japanese beech (*Quercus aliena*) and Japanese red pine, with a stand density of 2500/ha. To investigate the ^{137}Cs distribution at different canopy heights, a monitoring tower was built in each forest site. The heights of the monitoring towers and stories sampled are given in **Tables 1**. Fresh and dead leaf samples were collected at different heights in the canopy, and ^{137}Cs concentrations were determined for the different forest types at different time points (25 July and 18 November 2011).

Aerial dose rates and count rates of gamma rays emitted from ^{137}Cs were measured at different heights in the canopy during six periods between July 2011 and February 2012 using a portable germanium gamma ray detector (Detective-DX-100T; Ortech, Oak Ridge, Tennessee, USA). The width of the contribution area included all directions in front of the detector and the detection length was an almost infinite distance.

As a preliminary experiment, aerial dose rates were measured at different heights outside of the forest to investigate the relationship between the aerial dose rate and the distance from the ground surface. For this experiment, flat cultivated land with no forest within 200 m was selected.

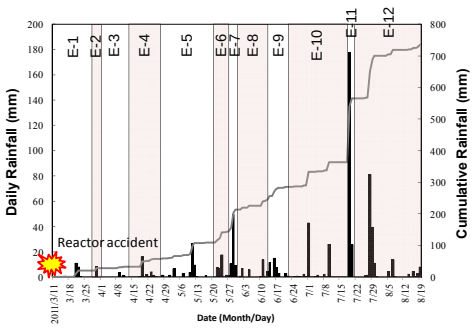

Figure 2. Daily rainfall and cumulative rainfall during the observation period.

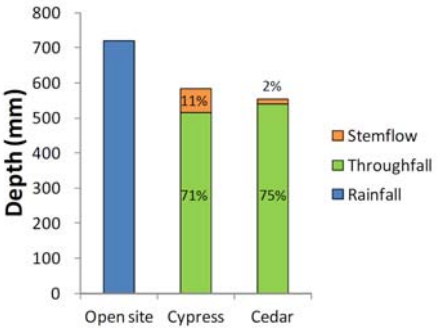

Figure 3. Proportions of throughfall and stemflow to open rainfall during the observation period.

3. Results and discussion

3.1. Total rainfall, throughfall, and stemflow at Tochigi

The total rainfall during the whole observation period was 740 mm at the Tochigi site. We observed 12 separate rainfall events with amounts ranging from 5.3 to 184.4 mm (**Figure 2**). The total amounts of throughfall and stemflow were 514 ± 71 mm and 70 ± 4 mm for the cypress forest, whereas the corresponding values for the cedar forest were 539 ± 81 mm and 16 ± 2 mm, respectively. Consequently, 71% and 75% of rainwater reached the forest floor via throughfall, and 11% and 2% of rainwater reached the forest floor via stemflow. The fractions of rainwater intercepted by the canopy were calculated as 18% and 23% for the cypress and cedar forests, respectively (**Figure 3**).

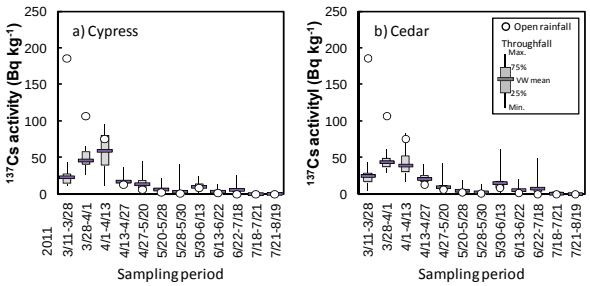

Figure 4. Cesium-137 concentrations in rainfall and throughfall.

3.2. Canopy interception of ^{137}Cs deposited at Tochigi

Temporal variations in ^{137}Cs concentrations in rainwater and throughfall are shown in **Figure 4**, in which the ranges and volume-weighted means of ^{137}Cs concentrations for the 20 throughfall collectors are plotted. The ^{137}Cs concentration in rainfall was highest during the first sampling period, while the concentration in throughfall was much lower than in rainfall. This finding indicates that large amounts of the deposited ^{137}Cs were intercepted during passage through the forest canopy. After mid-April, the concentration in throughfall tended to exceed that in open rainfall. This result indicates that removal of ^{137}Cs, rather than absorption onto the canopy, was occurring. Consequently, the apparent ^{137}Cs concentration in throughfall became higher than the original concentration in rainfall.

At the Tochigi site, the total atmospheric deposition of ^{137}Cs after the Fukushima reactor accident was 8026 Bq m^{-2}. Sixty-eight percent of the total ^{137}Cs fallout occurred during the first observation period, from 11 to 28 March (**Figure 5**). The atmospheric deposition of ^{137}Cs occurred in association with rainfall. The atmospheric input of ^{137}Cs decreased exponentially with time. However, deposition of ^{137}Cs onto the forest floor occurred mainly as a result of throughfall. Total ^{137}Cs depositions onto the forest floor during the whole observation period were 2912 Bq m^{-2} for the cypress stand and 2778 Bq m^{-2} for the cedar stand. In contrast, ^{137}Cs deposition via stemflow was one to two times lower than that of throughfall, with total depositions via stemflow for the cypress and cedar stands of 119 Bq m^{-2} and 28 Bq m^{-2}, respectively. The deposition rate, which represents the ratio of ^{137}Cs deposition onto the forest floor against total atmospheric deposition, is also shown in Figure 5.

For the period of March through mid-April 2011, more than 80% of the total deposited ^{137}Cs was intercepted and retained in the canopy in both coniferous forests. Leaf samples were collected from cypress and cedar trees on 12 May 2011 to confirm the retention of the deposited ^{137}Cs on plant surfaces. The ^{137}Cs concentrations in leaf samples collected from seven different heights in the canopies of the cypress and cedar stands ranged from 1.33 to 2.92 kBq kg^{-1} and 0.30 to 0.49 kBq kg^{-1}, respectively. The corresponding values in

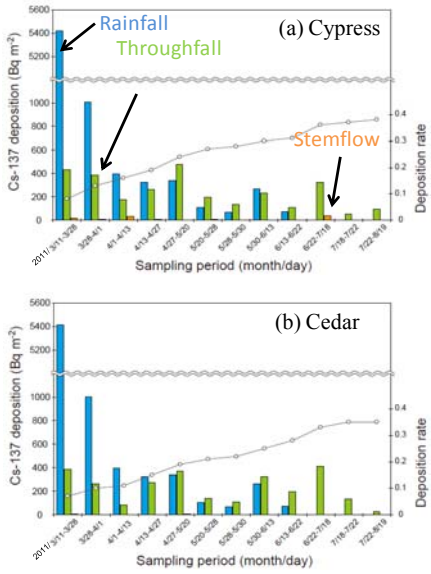

Figure 5. ^{137}Cs deposition via throughfall and stemflow during each observation period in (a) cypress and (b) cedar forests.

litter samples were 9.21 kBq kg^{-1} and 12.9 kBq kg^{-1}, respectively.

Cesium-137 concentrations in leaf samples were higher in the cypress forest than in the cedar forest. However, the amounts of total canopy interception by both forest types were fairly similar, suggesting that the difference in ^{137}Cs concentrations in leaves was derived from the differences in leaf density and canopy biomass between cypress and cedar forests. Nevertheless, the high concentrations in leaf samples indicated retention of the deposited ^{137}Cs on plant surfaces during the passage of contaminated rainwater through the forest canopy. Furthermore, the high ^{137}Cs concentrations in the litter samples suggested that the transfer of ^{137}Cs from the canopy to the forest floor increased the contamination of the litter layer.

3.3. Transfer of ^{137}Cs from the canopy to the forest floor at Tochigi

The deposition of ^{137}Cs onto the forest floor via throughfall decreased with the ^{137}Cs content in open rainfall. ^{137}Cs deposition in rainfall decreased exponentially with time, but the ^{137}Cs content in

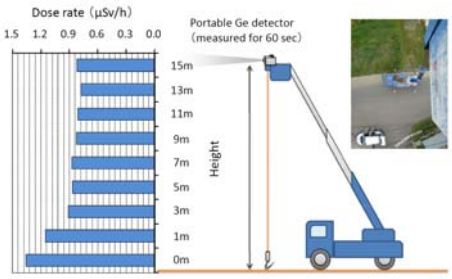

Figure 6. Vertical distribution of dose rate outside of the forest.

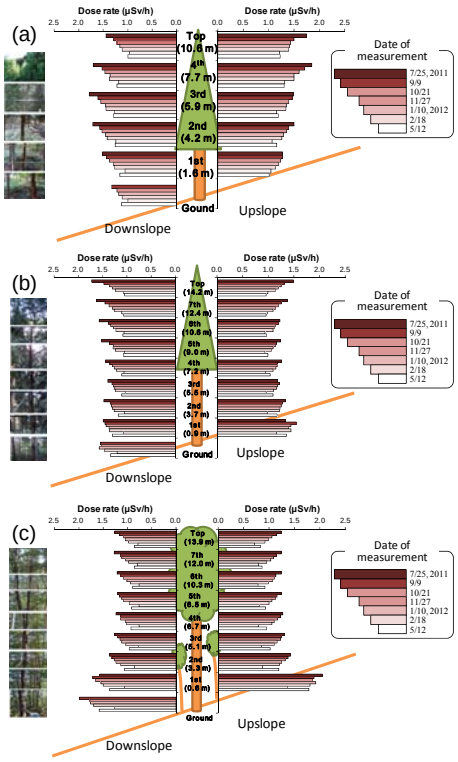

Figure 7. Vertical distribution of the aerial dose rate and temporal changes in each forest type.

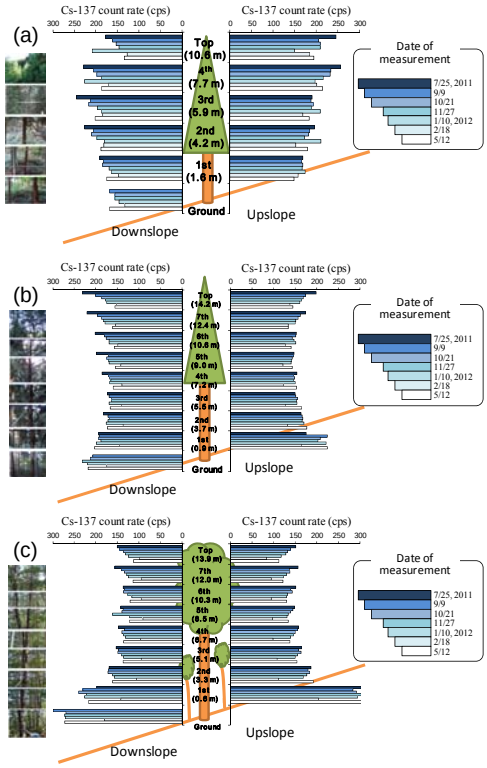

Figure 8. Vertical distribution of the ^{137}Cs count rate (cps) and temporal changes in each forest type.

throughfall decreased more slowly than that in rainfall. Cesium-137 deposition via throughfall tended to exceed the atmospheric input from rainfall after May. These data indicated that ^{137}Cs in the canopy was detached from the plant surface and subsequently transferred to the forest floor by rainwater. At the end of the observation period, the canopy interception ratios (the total amount of ^{137}Cs intercepted by the forest canopy against total atmospheric deposition) were calculated to be 62% for the cypress stand and 65% for the cedar forest.

Bunzl et al. [5] measured temporal changes in ^{137}Cs in the soil layer in an old spruce forest near Munich, Germany. The old spruce forest accumulated fallout from the Chernobyl reactor accident, with an estimated total ^{137}Cs deposition of 20 kBq m^{-2} after the accident. Approximately 600 days were required for all deposited ^{137}Cs to reach the forest floor, primarily via throughfall. For the cypress and cedar forests, 62% and 65% of atmospherically deposited ^{137}Cs was retained in the canopy 160 days after initial deposition. However, the corresponding value for the old spruce forest was 35.8%. Although the level of radioactive contamination in the old spruce forest was twice that in the cypress and cedar forests, the data suggested that the transfer of ^{137}Cs in a coniferous forest plantation in Japan was slower than in the old spruce stand.

3.4. Vertical distribution of aerial dose rate and ^{137}Cs count rate in cedar and broadleaved forests in Fukushima

Outside of the forest, the aerial dose rate decreased with increasing height up to 5 m from the ground surface, and thereafter remained almost constant at heights ≥ 5 m (**Figure 6**).The higher dose rate near the ground surface can be attributed to the accumulation of atmospherically deposited radionuclides in the surface soil. The influence of radiation from the ground surface became apparent constant above 5 m height because of the wider view of the detection area.

The vertical distributions of dose rate in the cedar and broadleaf forests are shown in **Figure 7**. In the young cedar forest, the highest aerial dose rate was found in the middle of the canopy. However, in the mature forest, the highest value was detected in the upper canopy and again displayed an increasing trend near the ground surface. The broadleaf forest had an almost uniform dose rate in the canopy; however, it increased toward the ground surface.

We measured the concentrations of ^{137}Cs in leaves at different heights and on different dates in each forest (**Table 1**). In summer 2011, ^{137}Cs concentrations were higher in the leaves of the cedar forest than in the leaves of the broadleaf forest. However, litter samples from the broadleaf forest contained significantly higher ^{137}Cs concentrations. In the evergreen cedar forest, atmospherically deposited ^{137}Cs may have been efficiently intercepted and retained by the forest canopy, whereas in the broadleaf stand, the deposited ^{137}Cs may have reached the forest floor because leaves had fallen at the time of atmospheric deposition. For all forest types, ^{137}Cs concentrations in leaf samples tended to decrease

Table 1. Cesium-137 concentrations in leaf samples.

Height (m)	2011/7/25 Fresh leaves ^{137}Cs act. (kBq/kg)	error	2011/7/25 Dead leaves ^{137}Cs act. (kBq/kg)	error	2011/11/18 Fresh leaves ^{137}Cs act. (kBq/kg)	error	2011/11/18 Dead leaves ^{137}Cs act. (kBq/kg)	error
(a) Young cedar								
10.6	97.5	3.0	no leaf	–	16.8	1.5	no leaf	–
7.7	28.5	1.6	33.8	1.8	10.4	1.6	19.6	1.8
5.9	33.7	1.6	33.3	2.3	20.1	2.0	8.8	1.4
4.2	44.1	2.8	67.6	3.7	no leaf	–	no leaf	–
1.6	no leaf	–	no leaf	–	no leaf	–	no leaf	–
Litter	–	–	77.4	3.2	–	–	81.1	3.9
Mean	50.9	2.3	44.9	2.7	15.8	1.7	14.2	1.6
(b) Mature cedar								
14.2	34.9	2.0	146.4	4.2	13.2	0.8	23.8	2.1
12.4	65.4	2.6	161.4	5.3	21.7	1.0	25.8	2.4
10.6	49.4	2.2	75.4	3.9	20.7	1.1	26.2	2.3
9.0	49.0	1.9	157.2	4.5	26.4	1.1	39.7	2.9
7.2	32.8	2.2	55.0	3.2	no leaf	–	43.4	2.6
5.5	no leaf	–	no leaf	–	no leaf	–	no leaf	–
3.7	no leaf	–	no leaf	–	no leaf	–	no leaf	–
0.9	no leaf	–	no leaf	–	no leaf	–	no leaf	–
Litter	–	–	99.4	2.8	–	–	15.7	1.5
Mean	46.3	2.2	119.1	4.3	20.5	1	31.8	2.5
(c) Broadleaf								
13.9	6.4	1.0	no leaf	–	4.6	0.5	5.5	0.6
12.0	16.4	0.2	no leaf	–	10.1	0.9	11.6	1.9
10.3	7.7	1.2	no leaf	–	10.2	0.8	9.5	1.0
8.5	10.3	1.4	no leaf	–	7.9	0.7	8.0	0.8
6.7	39.3	1.7	no leaf	–	9.4	1.0	8.7	1.4
5.1	7.0	0.7	no leaf	–	no leaf	–	no leaf	–
3.3	20.7	2.0	no leaf	–	no leaf	–	no leaf	–
0.6	no leaf	–	no leaf	–	no leaf	–	no leaf	–
Litter	–	–	413.1	6.7	–	–	8.6	0.6
Mean	15.4	1.3	–	–	8.5	0.8	8.6	1.2

between summer (25 July) and winter (18 November) in 2011. The reduction of ^{137}Cs concentrations in leaf samples can be attributed to the weathering of highly contaminated old leaves and the development of new leaves. The litter layers displayed marked decreases in ^{137}Cs concentrations in the mature and broadleaf stands, although these concentrations were expected to increase with time as the forest floor accumulated additional ^{137}Cs, which was removed from forest canopies. In the young cedar stand, ^{137}Cs concentrations in the litter layer were fairly similar in both periods.

With regard to the vertical distribution of ^{137}Cs in forest canopy, ^{137}Cs count rates clearly varied among tree species and ages (**Figure 8**). We compared the vertical distribution of ^{137}Cs count rates on six different days for each forest. In all forest types, the upper canopy displayed a marked decrease in the ^{137}Cs count rate with time, whereas the ground surface exhibited no change or a slight increase. This finding may indicate that the ^{137}Cs absorbed on leaves was moving downward through the forest canopy. As shown in Figure 8, the vertical distribution of ^{137}Cs in forest canopies can be determined by in-situ measurement of radioactivity using a portable germanium detector. Although further calibration procedures are necessary, in-situ measurement of radioactivity can be a useful tool for the monitoring of ^{137}Cs transfer in forest canopies.

4. Summary

In this study, we investigated the behavior of ^{137}Cs deposited in a forest environment immediately after the Fukushima reactor accident. More than 90% of the initial atmospheric deposition of ^{137}Cs was intercepted by cypress and cedar forest canopies, and 60% of the total deposition remained on the canopies 5 months after initial deposition. The transfer rate of ^{137}Cs from the canopy to the forest floor was slower than in an old spruce stand affected by fallout from the Chernobyl reactor accident. The high interception fraction of the deposited ^{137}Cs and the slow transfer rate from the canopy to forest floor in coniferous canopies indicated that the canopy can act as a secondary source of radioactive contamination to the forest floor. Furthermore, burning of contaminated wood can be a source of air contamination due to the resuspension of deposited ^{137}Cs. Therefore, the monitoring of long-term transfer of deposited ^{137}Cs from the canopy to the forest floor is required to assess the extent of the radioactive contamination of forest environments and ecosystems.

References

[1] C. Butler, Radioactivity spreads in Japan, *Nature* 471 (2011), pp.555-556.

[2] Ministry of Education, Culture, Sports, Science and Technology, Japan (MEXT), *Monitoring information of environmental radioactivity level*, http://radioactivity.mext.go.jp/en/, (2011).

[3] Y. Onda, T. Gomi, S. Mizugaki, T. Nonoda and R. Sidle, An overview of the field and modeling studies on the effects of forest devastation on flooding and environmental issues, *Hydrological Processes* 24 (2010), pp.527-534.

[4] H. Kato, Y. Onda and T. Gomi, Interception of the Fukushima reactor accident-derived ^{137}Cs, ^{134}Cs, and ^{131}I by coniferous forest canopies, *Geophysical Research Letters* 39, L20403 (2012), pp.1-6.

[5] K. Bunzl, W. Schimmack, K. Kreutzer and R. Schierl, Interception and retention of Chernobyl-derived ^{134}Cs, ^{137}Cs and ^{106}Ru in a spruce stand, *The Science of the Total Environment* 78 (1989), pp.77-87.

[6] F. O. Hoffman, K. M. Thiessen and R.M. Rael, Comparison of interception and initial retention of wet-deposited contaminants on leaves of different vegetation types, *Atmospheric Environment* 29, 15 (1995), pp.1771-1775.

[7] G. Pröhl and F. O. Hoffman, Radionuclide interception and loss processes in vegetation, IAEA-TECDOC-857 (1996), pp.9-44.

[8] R.P. Kinnersley, G. Shaw, J. N. B. Bell, M. J. Minski and A. J. H. Goddard, Loss of particulate contaminants from plant canopies under wet and dry conditions, *Environmental Pollution* 91, 2 (1996), pp.227-235.

[9] R.P. Kinnersley, A. J. H. Goddard, M. J. Minski and G. Shaw, Interception of caesium-contaminated rain by vegetation, *Atmospheric Environment* 31, 8(1997), pp.1137-1145.

[10] G. Pröhl, Interception of dry and wet deposited radionuclides by vegetation, *Journal of Environmental Radioactivity* 100 (2009), pp.675-682.