Movements and Storages of Radiocesium in a Forest Ecosystem in Fukushima

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The disaster that occurred at the Fukushima Daiichi Nuclear Power Plant in March 2011 resulted in a massive dispersion of radioactive cesium (¹³⁷Cs) over vast forests and other surrounding areas. The author has conducted intensive monitoring in a water catchment area located in northern Fukushima to determine and explain how exactly ¹³⁷Cs migrates to and builds up in a forest ecosystem before being discharged from it. This monitoring revealed that the amount of ¹³⁷Cs that is discharged from the forest over the course of one year was at least two orders of magnitude less than the estimated amount of deposition immediately after the disaster. It has been suggested that the migration takes place mainly in the form of suspended solids with particulate organics serving as important carriers. The largest ¹³⁷Cs pools in the forest ecosystem proved to be the litter layer and the topsoil. The circulation of ¹³⁷Cs was also indicated within flora, including tall trees. The dispersion of ¹³⁷Cs within the biological communities of animals and other creatures was more notable in terms of migration through food webs extending from animals that feed on fallen leaves and their fragments as compared to migration through food webs extending from animals that feed on live leaves. No increase in the ¹³⁷Cs concentration was observed with the rise in the trophic level, which demonstrates that no biological accumulation took place.

KEYWORDS: Fukushima, radioactive cesium, forest ecosystem, biogeochemical cycle, suspended solid, food web, biological accumulation, root absorption

I. Introduction

The accident that occurred at the Fukushima Daiichi Nuclear Power Plant in March 2011 resulted in a massive dispersion of radioiodine (approx. 1.5×10^{17} Bq of 131 I) and radioactive cesium (approx. 1.2×10^{16} Bq of 137 Cs) in Fukushima and its surrounding areas ¹). The forest coverage exceeds 70 percent in most municipalities within these areas. The deposited radioactive materials raise concerns in terms of exposure in human habitats, damage to the forestry and forest product industries, and impairment of water sources in forests.

The initial survey, which was led by the Ministry of Education, Culture, Sports, Science

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and Technology (MEXT) and the Ministry of Agriculture, Forestry and Fisheries (MAFF) soon after the deposition began in FY2011, indicated that the radioactive cesium that fell on forests was retained on the crowns of evergreen trees and in the litter layer around deciduous trees ^{1, 2)}. It has been indicated that clay minerals in soil strongly adsorb radioactive cesium ³⁾. It has also been reported that radioactive cesium is discharged into streams and rivers along with soil particles due to any soil erosion and runoff (e.g., Wakiyama et al., 2010)⁴⁾.

In a forest ecosystem, the radioactive cesium deposited on tree crowns migrates toward the forest floor over time by means of eluviation caused by rain⁵⁾ or defoliation^{6,7)}. Hashimoto et al.²⁾ conducted a numerical simulation based on data obtained up to 2012, and they predicted that most of the radioactive cesium that was deposited on the tree crowns would reach the forest bed within the first five years.

Dissolved radioactive cesium is absorbed by microbes, algae, plants, and various other creatures in a forest ecosystem. In a biological community, the radioactive cesium captured by algae, plants, and other primary producers is most likely passed along a food web to a wide range of creatures. Ultimately, it is likely to move up the trophic levels to fish, birds, and mammals. Many past studies on the migration of radioactive substances through food webs have attempted to determine whether biological accumulation takes place^{8,9}.

To deal with forest contamination in the affected areas, it was considered essential to clarify in detail how radioactive cesium deposited in forests is redistributed by migration within the ecosystem and how much of the radioactive cesium is discharged from the system in the initial years. Accordingly, the author and his colleagues conducted a survey in a forest located in northern Fukushima with the following aims: 1) to determine the redistribution mechanism for radioactive cesium in the forest; 2) to assess the amount of radioactive cesium migration among creatures in the hydrological process; and 3) to monitor radioactive cesium migration among creatures in the food web of the biological community. This paper reports the survey results using data obtained by the end of FY2014 to consider necessary surveys and measures for the future. It may be noted that most of the findings have already been published in the references¹⁰⁻¹².

II. Survey Method

A field survey was conducted at the gully head of the Kami-Oguni River, which runs through the Kami-Oguni district of Ryozenmachi, Date City, in northern Fukushima. According to aircraft observations conducted in June and July of 2013, the air dose rate in the surrounding area ranged from 1.0 to $1.9 \,\mu\text{Sv} \,\text{h}^{-1}$ and the expected total amount of ¹³⁷Cs precipitation was between 100 and 300 kBq m^{-2 13}. The main parts of the survey site are covered by a mixed secondary forest made up of Japanese red pine (*Pinus densiflora*) and deciduous broadleaf trees, such as jolcham oak (*Quercus serrata*) and Japanese elm (*Zelkova serrata*). An approximately 50-year-old artificial forest of Japanese cedar (*Cryptomeria japonica*) extends along the valley.

To track the flux from the radioactive cesium that migrates along with water in these forests, a hydrological observation was conducted to measure the radioactive cesium concentration at various stages from the precipitation to the runoff (e.g., precipitation, passage through crowns, and spillover into streams). Two square plots were assigned to the mixed forest of deciduous broadleaf trees and Japanese red pine as the main part of the forest system. Another plot was assigned to the artificial forest of Japanese cedar. In each of these plots, the litterfall (i.e., fallen leaves and branches in a forest) was sampled, and measurements were taken to determine the amount and concentration of radioactive cesium in the throughfall and stemflow.

In addition, land and aquatic creatures were sampled along the stream to determine the extent to which radioactive cesium is transmitted within the biological community. The sampled creatures were identified before the concentration of radioactive cesium in their tissues was measured.

Moreover, standing trees in the main forest were cut down and sampled in November of both 2012 and 2013 to estimate the amount of radioactive cesium buildup above the ground. The samples were divided into live leaves, branches, and trunks (bark, sapwood, and heartwood) to measure the radioactive cesium concentration^{10, 11}.

III. Results and Discussion

1. ¹³⁷Cs Concentration in Plants

Live leaves on evergreen Japanese cedar can last for about three years. The leaves that foliate in the current year are called "current leaves," while other leaves that foliated earlier are called "older leaves." Presumably, a certain proportion of the leaves were still attached in the years that followed the deposition of radioactive cesium on them in March 2011. In 2012, the ¹³⁷Cs concentration in live leaves exceeded 10,000 Bq kg⁻¹ for both current leaves and older leaves. The concentration measured in 2013 had dropped to 3,500 Bq kg⁻¹ in older leaves and 2,700 Bq kg⁻¹ in current leaves (**Figure 1**). The similar concentration levels observed between leaves that foliated in 2012 and leaves that remained from the previous year suggest that the deposited radioactive cesium was translocated from the crown or other parts of the trees to newly formed leaves. This means that radioactive cesium on the surface of leaves, branches, and trunks can seep into the tree body and that it can be carried via nutrient translocation inside the tree body. The decline in the ¹³⁷Cs concentration that was observed with older leaves in 2013 can probably be explained by them being replaced with new leaves that have a relatively low concentration and rainfall washing away some of the radioactive cesium.

As a deciduous tree, jolcham oak foliates in early summer and defoliates in late fall, which means that live leaves on the crown are replaced every year. The ¹³⁷Cs concentration in live leaves was around 1,000 Bq kg⁻¹ in both 2012 and 2013. In March 2011, when radioactive cesium first fell on the forest, live leaves had not foliated yet. The ¹³⁷Cs in these samples seems to have seeped into the tree body from the surface of the trunks and branches before further translocation. Meanwhile, some of the ¹³⁷Cs that was deposited on fallen leaves and the like on the forest floor was absorbed through roots before being transferred to new leaves.



Figure 1 ¹³⁷Cs concentration in live leaves on jolcham oak and Japanese cedar

The samples were taken by cutting down standing trees in November of both 2012 and 2013. The concentration was measured separately for older leaves and current leaves on Japanese cedar (Ohte et al., 2015¹¹).



Figure 2 ¹³⁷Cs concentration in the sapwood and heartwood of Japanese cedar and jolcham oak The samples were taken by cutting down standing trees in November of both 2012 and 2013 (Ohte et al., 2015¹¹).

The 137 Cs concentration exceeded 10,000 Bq kg ${}^{-1}$ in most bark samples from jolcham oak during the same period in 2012 11 .

The marginal difference in concentration between the sapwood and the heartwood of Japanese cedar as compared to jolcham oak in both 2012 and 2013 (**Figure 2**) indicates a much faster translocation of radioactive cesium in a tree trunk of Japanese cedar.

The above findings demonstrate the active movement of radioactive cesium via the nutrient translocation mechanism of trees. Especially, the discovery that a high concentration of radioactive cesium in bark migrates to sapwood and then translocates to leaves is important. The next task would be to conduct a quantitative measurement of the ¹³⁷Cs absorption through roots under a forest floor covered in heavily contaminated leaves.

2. Migration of Radioactive Cesium from Tree Crowns to the Forest Floor

Among the three plots, the greatest migration of ¹³⁷Cs from tree crowns to the forest floor via litterfall was observed in the artificial cedar forest (**Table 1**) ¹⁴. As explained in the previous section, this is probably due to the larger amount of radioactive cesium that was deposited on evergreen tree crowns. Even when the throughflow and stemflow were taken into consideration in addition to migration via litterfall, the amount of migration was found to be greatest in the artificial forest of evergreen cedar.

The migration from the tree crowns to the forest floor supplies ¹³⁷Cs to microbes in the litter and humus layers as well as to the plants that extend their roots there. However, the availability of ¹³⁷Cs for microbes and plants is believed to be quite different between migration via litterfall and migration via throughfall and stemflow.

In addition to confirming the amount absorbed by trees through their roots as mentioned earlier, detailed surveys need to be conducted to determine the standing stock of radioactive cesium in the upper-litter and humus layers that is readily absorbable by plants and microbes as well as other factors such as seasonal changes in the standing stock of radioactive cesium.

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	Annual average ¹³⁷ Cs concentration (Bq L ⁻¹ : Throughfall and stemflow; Bq kg ⁻¹ : Litterfall)			¹³⁷ Cs flux (Bq m ⁻² year ⁻¹)		
	DP1	DP2	СР	DP1	DP2	СР
Throughfall	3.10	3.01	5.54	3,254	1,694	3,388
Stemflow	4.01	0.97	2.16	458	101	69
Litterfall	8,068	7,464	17,887	2,904	2,125	7,518

Table 1 Annual average ¹³⁷Cs concentration and annual ¹³⁷Cs flux for throughfall, stemflow, and litterfall

Footnote: DP1: Mixed forest of deciduous broadleaf trees and Japanese red pine trees 1; DP2: Mixed forest of deciduous broadleaf trees and Japanese red pine trees 2; CP: Artificial forest of Japanese cedar. (Original data source: Endo et al., 2015¹⁴)



Figure 3 Precipitation, concentration of suspended solids, concentration of ¹³⁷Cs, and river flow over time during a flooding event on October 15, 2013 (Iseda, 2015¹⁵)

3. Radioactive Cesium Runoff into Streams

Figure 3 presents changes in the concentrations of suspended solids and ¹³⁷Cs over time during their rain-induced runoff into a swollen stream in October 2013 (Iseda, 2015)¹⁵). These changes are almost synchronized, which indicates that suspended solids served as important carriers for ¹³⁷Cs runoff.

The annual ¹³⁷Cs runoff from the catchment area was estimated by taking into consideration the changes in ¹³⁷Cs concentration associated with changes in the stream's flow rate. The estimated amount for the period from August 31, 2012, to August 30, 2013, was 330 Bq m⁻² year^{-1 15)}. However, it should be noted that just one major flood caused by heavy rain in mid-October 2013 caused a ¹³⁷Cs runoff of 227 Bq m⁻² in a matter of a few days. Given this, it is quite important to observe flooding to track the ¹³⁷Cs runoff from the catchment area accurately.

As mentioned earlier, the estimated ¹³⁷Cs deposition in this area is 100 to 300 kBq m⁻², which is three orders of magnitude greater than the estimated runoff in a period of one year.

Given ¹³⁷Cs's half-life of roughly 30.1 years, the amount of radioactive cesium that is discharged from a forest ecosystem into rivers through hydrological processes is apparently much less than the amount that disappears due to radioactive decay within the system.

4. Migration of Radioactive Cesium in Food Webs

Figure 4 presents the ¹³⁷Cs concentration among samples of land and aquatic creatures broken down by functional group. The concentration for fallen leaves, fungi, scavengers, and predators was significantly higher than that for the live leaves on plants and plant-eating creatures.

Fallen leaves and their fragments that have built up on the ground surface retain the largest amount of ¹³⁷Cs, which migrated noticeably among land creatures from these sources. Fallen leaves and their fragments and benthic algae, which serve as basic food for aquatic creatures, had ¹³⁷Cs concentration levels that were somewhere between the concentration levels for live leaves and those for fallen leaves on the ground. The ¹³⁷Cs concentrations among creatures in higher trophic levels could be explained by a combination of the concentration levels explained earlier ¹⁶.

The nitrogen stable isotope ratio increases in the tissues of creatures in higher trophic levels, so it can be regarded as a relative indicator of tropic levels. The measured ratio and the ¹³⁷Cs concentration among creature samples exhibited a slightly negative correlation. In other words, the ¹³⁷Cs concentration was lower among creatures in higher trophic levels, which indicates that no biological accumulation of ¹³⁷Cs took place in the biological community studied in this survey ¹⁶.



Figure 4 ¹³⁷Cs concentration by functional group

The alphabetic denotation assigned to each plot represents its functional group according to the statistical grouping. The same letter represents the same functional group. There were samples below detection limit for predators and consumers in aquatic, plant-eating creatures and predators in land. The number of those samples is indicated after circle (Murakami et al. 2014¹⁶).



Figure 5 Distribution of the standing stock of ¹³⁷Cs among parts of the plots covered respectively by a deciduous broadleaf forest and an artificial cedar forest as of September 2014 (yet to be published)
Layer O represents a sedimentary layer of fallen leaves and branches. Layers A1 and A2 are the most superficial mineral soil layers with a notable comingling of organic matter.

IV. Conclusions

Figure 5 presents the standing stock of ¹³⁷Cs in each part of the plots covered respectively by a deciduous broadleaf forest and an artificial cedar forest as of September 2014.

These different types of forests commonly retain the greatest amount of ¹³⁷Cs in their litter layers and topsoil. The buildup inside plants is expected to be relatively small. Nonetheless, the most crucial finding of the monitoring that has been conducted so far is the fact that radioactive cesium continues to migrate actively without any stable distribution with a specific spatial alignment. The internal circulation in the ecosystem was particularly visible along nutrient cycle pathways between plants and the soil. The ¹³⁷Cs migration diminished year by year in the evergreen artificial forest of Japanese cedar, with the concentration in the trees' new needle leaves at the level of a few thousand Bq kg⁻¹. In the future equilibrium, the amount of absorption into the tree body is expected to reach a similar level to the amount of migration to the forest floor.

Meanwhile, a certain portion of the ¹³⁷Cs is believed to seep down into the mineral soil layer to be retained by clay minerals. However, internal circulation between plants and the litter and humus layers is expected to last for a long time. The availability of radioactive cesium for creatures is reduced by its adsorption and retention by clay minerals. However, it is reasonable to assume that radioactive cesium remains available for creatures as long as the internal circulation mechanism and the circulated amount of radioactive cesium must be observed carefully and continuously. In addition to the monitoring that is to be continued in the medium to long term, detailed studies on the processes that take place in litter layers and on the ground surface remain important.

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