

# Chapter 14

## Ecosystem Monitoring of Radiocesium Redistribution Dynamics in a Forested Catchment in Fukushima After the Nuclear Power Plant Accident in March 2011

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**Abstract** The accident at the Fukushima Daiichi Nuclear Power Plant in March 2011 emitted  $1.2 \times 10^{16}$  Bq of cesium-137 ( $^{137}\text{Cs}$ ) into the surrounding environment. Radioactive substances, including  $^{137}\text{Cs}$ , were deposited onto forested areas in the northeastern region of Japan.  $^{137}\text{Cs}$  is easily adsorbed onto clay minerals in the soil; thus, a major portion of  $^{137}\text{Cs}$  can be transported as eroding soil and particulate organic matter in water discharge. Dissolved  $^{137}\text{Cs}$  can be taken up by microbes, algae, and plants in soil and aquatic systems. Eventually,  $^{137}\text{Cs}$  is introduced into insects, worms, fishes, and birds through the food web. To clarify the mechanisms of dispersion and export of  $^{137}\text{Cs}$ , within and from a forested ecosystem, we conducted intensive monitoring on the  $^{137}\text{Cs}$  movement and storage in a forested headwater catchment in an area approximately 50 km from the Nuclear Power Plant. Two major pathways of  $^{137}\text{Cs}$  transport are as follows: (1) by moving water via dissolved and particulate or colloidal forms and (2) by dispersion through the food web in the forest-stream ecological continuum. The  $^{137}\text{Cs}$  concentrations of stream waters were monitored. Various aquatic and terrestrial organisms were

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periodically sampled to measure their  $^{137}\text{Cs}$  concentrations. The results indicate that the major form of exported  $^{137}\text{Cs}$  is via suspended matter. Particulate organic matter may be the most important carrier of  $^{137}\text{Cs}$ . High water flows generated by a storm event accelerated the transportation of  $^{137}\text{Cs}$  from forested catchments. Estimation of  $^{137}\text{Cs}$  export from the forested catchments requires precise evaluation of the high water flow during storm events. The results also suggested that because the biggest pool of  $^{137}\text{Cs}$  in the forested ecosystem is the accumulated litter and detritus,  $^{137}\text{Cs}$  dispersion is quicker through the detritus food chain than through the grazing food chain.

**Keywords** Cs • Forest ecosystem • Suspended solid • Food web • Bioaccumulation

## 14.1 Introduction

Following the Fukushima Daiichi Nuclear Power Plant accident in March 2011, approximately  $1.5 \times 10^{17}$  Bq of iodine-131 ( $^{131}\text{I}$ ) and  $1.2 \times 10^{16}$  Bq of cesium-137 ( $^{137}\text{Cs}$ ) were emitted into the surrounding environment (Ministry of Education, Culture, Sports, Science and Technology and Ministry of Agriculture, Forestry and Fisheries 2012). These radioactive substances were deposited on the northeastern region of Honshu Island, Japan. Forests occupy more than 70 % of the total area in these regions and are particularly important for local societies, not only because of their association with the forestry industry but also for their residential environment, which includes source areas of drinking water. The first phase of government surveys revealed that a major portion of the deposited radiocesium was trapped in the canopy and in the litter layer on the forest floor (Ministry of Education, Culture, Sports, Science and Technology; Ministry of Agriculture, Forestry and Fisheries 2012; Hashimoto et al. 2012). Radiocesium has been shown to be easily adsorbed by clay minerals and soil organic matter (Kruyts and Delvaux 2002), which can be transported by eroded soil, particulates, and dissolved organic matter through hydrological channels, streams, and rivers (e.g., Fukuyama et al. 2005; Wakiyama et al. 2010). Within the forested ecosystem, radioactive materials deposited on the tree canopies subsequently move to the forest floor by precipitation (Kinnersley et al. 1997; Kato et al. 2012) and litter fall (Bunzl et al. 1989; Schimmack et al. 1993; Hisadome et al. 2013). It is expected that the movement of radiocesium from the canopy to the forest floor will gradually decrease (Hashimoto et al. 2013), and it is considered that its movement in the Fukushima forests has been active because only 3.5 years have passed since the accident.

Dissolved radiocesium, which is relatively free from soil adsorption, can also be taken up by microbes, algae, and plants in soil and aquatic ecosystems. By propagating through the food web in the forest ecosystem, it was expected that radiocesium would eventually be introduced into soil, insects, worms, fishes, and birds. Many previous reports on the distribution and transfer of radionuclides have focused on bioaccumulation and the transition between trophic levels (Kitchings et al. 1976; Rowan and Rasmussen 1994; Wang et al. 2000).

Therefore, it is necessary to investigate the major pathways of radiocesium transfer in the forest, such as physical transportation via hydrological processes and biological transfer through the food web in terrestrial and aquatic ecosystems. To describe the current status and to examine the hypothesized mechanisms mentioned above, catchment scale biogeochemical observations have been conducted in the northern part of the Fukushima prefecture since August 2012 (Ohte et al. 2012, 2013), and they currently continue (February 2015). Here, we report the latest results and discuss the future perspectives for monitoring radiocesium levels in Fukushima forests.

## 14.2 Materials and Methods

### 14.2.1 Study Site

The study area is the Kami-Oguni River catchment, which is located approximately 50 km from the Fukushima Daiichi Nuclear Power Plant. The Ministry of Education, Culture, Sport, Science and Technology (2013) used aircraft survey devices to determine that the air dose rate in this region was 1.0–1.9  $\mu\text{Sv h}^{-1}$  and the total deposition rate of  $^{137}\text{Cs}$  was 300–600  $\text{kBq m}^{-2}$  in 2013. To intensively monitor the radioactive cesium dynamics, we set a small sub-catchment in a forest ecosystem in the headwater part of the Kami-Oguni River catchment, known as the Sanbo Observational Forest Catchment.

The geology of the catchment is dominated by volcanic rocks (andesite and basalt) formed by volcanic eruptions during the Miocene Epoch. The soil is classified as Dystrudepts (Soil Survey Staff 2014), which is characterized by high exchangeable  $\text{Mg}^{2+}$  and low  $\text{K}^{+}$  concentrations. The soil texture in the topsoil (A-Horizon) of this area was a mixture of clay < 0.002 mm (25.3 %), silt 0.02–0.002 mm (40.1 %), sand 2–0.02 mm (34.6 %) determined with the sieving method (Kato et al. unpublished data).

Major areas of Sanbo Observational Forest Catchment are covered by typical secondary stands that consist of broadleaf deciduous species dominated by *Quercus serrata* Murray and *Zelkova serrata*. Some parts of these secondary stands were mixed with naturally regenerated Japanese red pine (*Pinus desiflora*). The valley areas of the catchment were used as Japanese cedar (*Cryptomeria japonica*) plantations for timber production. *Pinus desiflora* and *Cryptomeria japonica* are evergreen conifers.

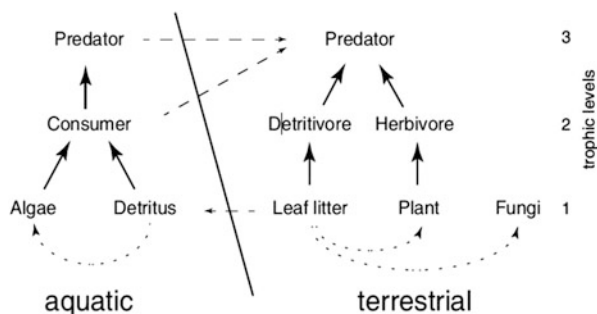
## 14.2.2 Field Observations and Sampling

To monitor the fluxes of radiocesium transported by water flow into and out of the forest catchment, we measured water fluxes and radiocesium concentrations for each elemental hydrological process, such as rainfall, throughfall, stemflow, and stream water discharge. The water discharge from the catchment was continuously measured using a partial flume with a water level recorder. Stream water was sampled monthly and intensively during storm events.

Three rectangular plots including two deciduous-pine mixed stands (DP1 and DP2, 20 m × 20 m) and one cedar plantation (CP, 10 m × 40 m) were selected to investigate the spatial distributions and movements of radiocesium in hydrological and biogeochemical processes within the plant and soil system. We conducted monthly samplings of litter fall, throughfall, stemflow, and soils from each plot, and quantified all samples. Details of the sampling are described in Endo et al. (2015).

Food web components including terrestrial and aquatic organisms in the continuum of forest and stream ecosystems were sampled to investigate the current status of radiocesium flows and storages affected by biological activities. All samples were collected from the study site and identified to the genus and species level, and then classified into 10 functional groups according to the criteria shown in Fig. 14.1. The sampling points for each sample were selected randomly within the study catchment. The samples were collected in May, July, and September 2012 and in February 2013. Detailed descriptions of the sampling and data analysis are presented in Murakami et al. (2014).

Whole trees were sampled in November 2012 and 2013 to estimate the stocks of radiocesium in the above ground biomass of the dominant trees, *Quercus serrata* and *Cryptomeria japonica*, at the study site. *Quercus serrata* is the most common species in the secondary deciduous forest in this region, and *C. japonica* is the most common species in the plantations for timber production in Japan. Three



**Fig. 14.1** Schematic food web of the present study. Ten functional groups were used as sampling units. *Solid lines* indicate trophic interactions and *dashed lines* indicate nutrient flow. *Broken lines* show spatial movements; e.g., transformation of tadpoles to frogs and dragonflies or supply of leaf litters from forest to stream (Murakami et al. 2014)

individuals of *Q. serrata* and one *C. japonica* were sampled at each sampling event in November of 2012 and 2013. Only a single *C. japonica* was sampled, because these trees were planted at a specific time and thus their tree sizes were homogeneous.

Radiocesium concentrations ( $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ ) of leaves, branches, and stems were measured separately. Moreover, stem samples were separated into sapwood and heartwood. Prior to these samplings and measurements, surveys of individual tree sizes were performed for three rectangle plots. All trees with a diameter at breast height (DBH)  $>5$  cm were measured for their DBH and height at the top of the canopy.

### 14.2.3 Sample Analysis

Germanium semiconductor detectors were used to measure radiocesium concentrations for all samples. Gamma-ray spectrometry was conducted using germanium detectors (Seiko EG&G). The measured values were corrected for the sampling day, and were expressed in  $\text{Bq kg}^{-1}$  of dry weight for the organic samples and in  $\text{Bq L}^{-1}$  for the water samples, respectively.

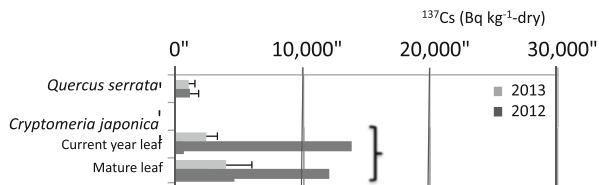
To evaluate the relative trophic levels for the sampled organisms, the stable nitrogen isotope ratio ( $\delta^{15}\text{N}$ ) for each sample was measured using SerCon ANCA GSL elemental analyzer interfaced to a SerCon Hydra 20–20 continuous flow isotope ratio mass spectrometer. Analysis and data treatments are explained in Murakami et al. (2014).

## 14.3 Results and Discussion

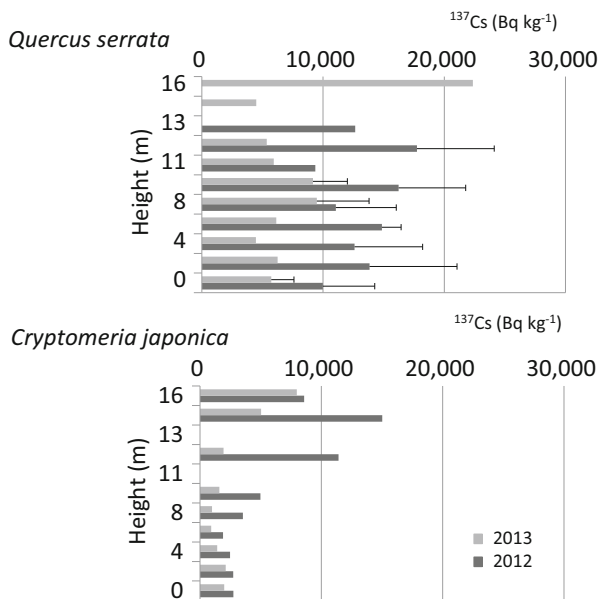
### 14.3.1 $^{137}\text{Cs}$ Concentrations of Tree Parts

The leaf  $^{137}\text{Cs}$  concentrations for *C. japonica* were higher than  $10,000 \text{ Bq kg}^{-1}$  for the mature and current leaves, but they decreased to  $3500 \text{ Bq kg}^{-1}$  for mature leaves and  $2700 \text{ Bq kg}^{-1}$  for current leaves (Fig. 14.2). These results indicate that the leaves that were on the canopy when the radiocesium was deposited still contained very high  $^{137}\text{Cs}$  levels in November 2012, and even the new leaves in 2012 had similarly high levels of  $^{137}\text{Cs}$  concentration. These results suggest that  $^{137}\text{Cs}$  was translocated from the old leaves to the new leaves. The decreasing  $^{137}\text{Cs}$  concentrations in the mature leaves implies that they were replaced with new leaves with lower  $^{137}\text{Cs}$  concentrations, and also that the  $^{137}\text{Cs}$  attached to the leaf surfaces of the old leaves was gradually washed out by rainfall.

On the other hand, leaves of *Q. Serrata* had 1000 and 990  $\text{Bq kg}^{-1}$  in 2012 and 2013, respectively. Considering that no living leaves were on the canopies of these



**Fig. 14.2** The <sup>137</sup>Cs concentration in living leaves of *Quercus serrata* and *Cryptomeria japonica*. Samples were collected from whole tree sampling conducted in early November 2012 and 2013. Because *C. japonica* is an evergreen species, it has multiple aged leaves, while *Q. serrata* is a deciduous species. Data without error bars were derived from the samples without replications



**Fig. 14.3** The <sup>137</sup>Cs concentration in the trunk bark of *Quercus serrata* and *Cryptomeria japonica*. Samples were collected from whole tree sampling conducted in early November 2012 and 2013. Data without error bars were derived from the samples without replications

deciduous trees when the radiocesium fell in March 2011, the <sup>137</sup>Cs in the living leaves was probably transported from the other parts of the tree in 2012 and 2013.

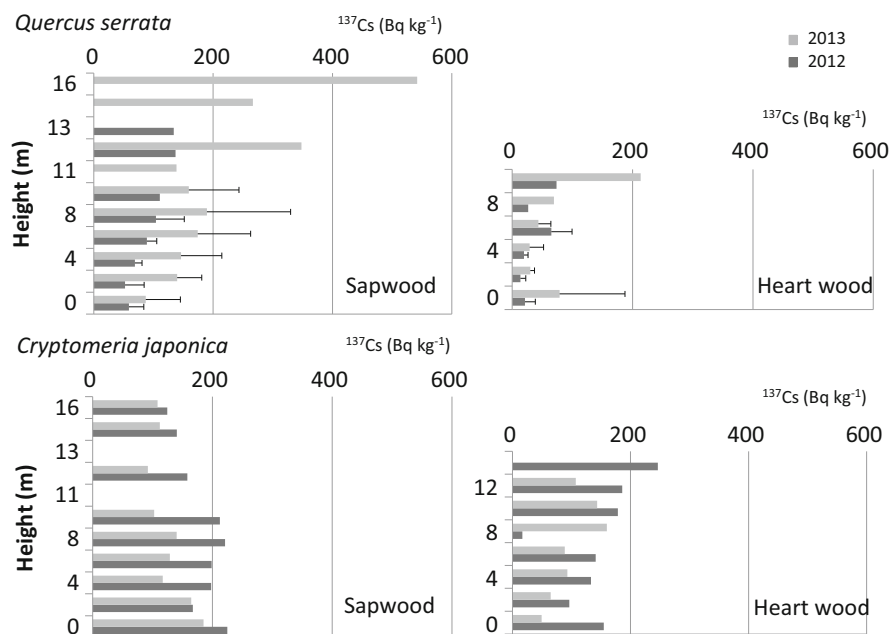
For *C. japonica*, the concentrations of <sup>137</sup>Cs in the bark within the evergreen canopy part of the tree (>9000 Bq kg<sup>-1</sup> in 2012 and 5000–7000 Bq kg<sup>-1</sup> in 2013) were higher than in the bark beneath the canopy (2000–5000 Bq kg<sup>-1</sup> in 2012 and 1000–2000 Bq kg<sup>-1</sup> in 2013; Fig. 14.3). In contrast, no significant vertical patterns of <sup>137</sup>Cs concentrations was found in the bark of *Q. serrata* (10,000–18,000 Bq kg<sup>-1</sup>), which did not have a canopy of leaves when the radiocesium was deposited in March 2011. This suggests that aerosols containing radiocesium deposited and adhered to the whole surface of tree trunks. The bark <sup>137</sup>Cs

concentration decreased to 5000–10,000 Bq kg<sup>-1</sup> except for the top part of the canopy in 2013. For both the evergreen conifer and deciduous trees, the decrease in <sup>137</sup>Cs concentration between 2012 and 2013 implies that the attached radiocesium on the trunk surfaces was leached out by rainwater, and was translocated into other part of the body. Removed with bark abrasion was also possible.

The <sup>137</sup>Cs concentration in *C. japonica* wood was 100–210 Bq kg<sup>-1</sup> for sap wood and 10–220 Bq kg<sup>-1</sup> for heartwood, and that of *Q. serrate* was 60–120 Bq kg<sup>-1</sup> for sap wood and 20–80 Bq kg<sup>-1</sup> for heartwood (Fig. 14.4). For the coniferous species *C. japonica*, the difference in <sup>137</sup>Cs concentrations was small between sapwood and heartwood, indicating that the translocation and/or dispersion occurred quickly within the wood.

While the <sup>137</sup>Cs concentration in *C. japonica* wood decreased between 2012 and 2013, the concentration increased for *Q. serrate* wood. This suggests that translocation from the bark, and probably from surface soils including litters, through roots to sapwood and heartwood might be delayed in *Q. serrate* compared to *C. japonica*, or might be greater in *Q. serrate* than *C. japonica*.

All of the results indicate that <sup>137</sup>Cs moved actively via nutrient transportation mechanisms and abiotic dispersion into the tree body. In addition to the movement



**Fig. 14.4** The <sup>137</sup>Cs concentrations in sapwood and heartwood of *Quercus serrata* and *Cryptomeria japonica*. Samples were collected from whole tree sampling conducted in early November 2012 and 2013

**Table 14.1** Annual average  $^{137}\text{Cs}$  concentration of throughfall, stemflow and litterfall, and the estimates of annual  $^{137}\text{Cs}$  flux by the different paths

	Annual mean $^{137}\text{Cs}$ concentration			$^{137}\text{Cs}$ flux		
	DP1	DP2	CP	DP1	DP2	CP
			( $\text{Bq L}^{-1}$ )			( $\text{Bq m}^{-2} \text{ year}^{-1}$ )
Throughfall	3.10	3.01	5.54	3254	1694	3388
Stemflow	4.01	0.97	2.16	458	101	69
			( $\text{Bq kg}^{-1}$ )			
Litterfall	8068	7464	17,887	2904	2125	7518

The plot codes; DP1, DP2 and CP indicate the deciduous-pine mixed stand #1, the deciduous-pine mixed stand #2 and the cedar plantation, respectively (calculated from the original data in Endo et al. 2015)

from bark (which continued to have high  $^{137}\text{Cs}$  concentrations) to sapwood, translocation to the juvenile leaves was clearly evident. It is currently difficult to evaluate the relative contributions to  $^{137}\text{Cs}$  in the wood from root uptake and translocation from the bark under field conditions. The root uptake rate of the  $^{137}\text{Cs}$  is the most important factor to be evaluated precisely and urgently.

### 14.3.2 $^{137}\text{Cs}$ Movement from the Canopy to the Forest Floor

The annual  $^{137}\text{Cs}$  fluxes from the canopy to the forest floor via throughfall, stem flow, and litter fall were summarized from October 2012 to September 2013 in Table 14.1. The annual  $^{137}\text{Cs}$  flux with litter fall was 2.9, 2.1, and 7.5  $\text{kBq m}^{-2} \text{ year}^{-1}$  for the DP1, DP2, and CP plots, respectively. The largest  $^{137}\text{Cs}$  flux in the CP plot can be explained by the highest  $^{137}\text{Cs}$  concentration in the litter from *C. japonica*.

The annual  $^{137}\text{Cs}$  fluxes for the sum of throughfall and stem flow were 3.7, 1.8, and 4.0  $\text{kBq m}^{-2} \text{ year}^{-1}$  for the DP1, DP2, and CP plots, respectively. The rate values were in the same order as those for litter fall. The total  $^{137}\text{Cs}$  flux by these pathways were 6.6, 3.9, 11.5  $\text{kBq m}^{-2} \text{ year}^{-1}$  for the DP1, DP2, and CP plots, respectively.

Each of these fluxes changed temporally depending upon phenology, seasonal variation of precipitation rate, and frequency of storm events (the data were not shown. See Endo et al. 2015). The  $^{137}\text{Cs}$  flux with litter fall was largest in late fall (late October to mid-November) while that with throughfall and stem flow was high in the middle of summer (July to September) when the precipitation rate was greatest. The  $^{137}\text{Cs}$  movement from the canopy to the forest floor occurred actively through the plant-growing season, even 2 years after the radiocesium was deposited.

The  $^{137}\text{Cs}$  availability for plants, algae, and microbes might be different between those attached to litters and those dissolved or suspended in throughfall or stem flow water. In order to understand the current mechanisms of radiocesium cycling between plants and soils, it is important to quantify the bioavailable  $^{137}\text{Cs}$  in the

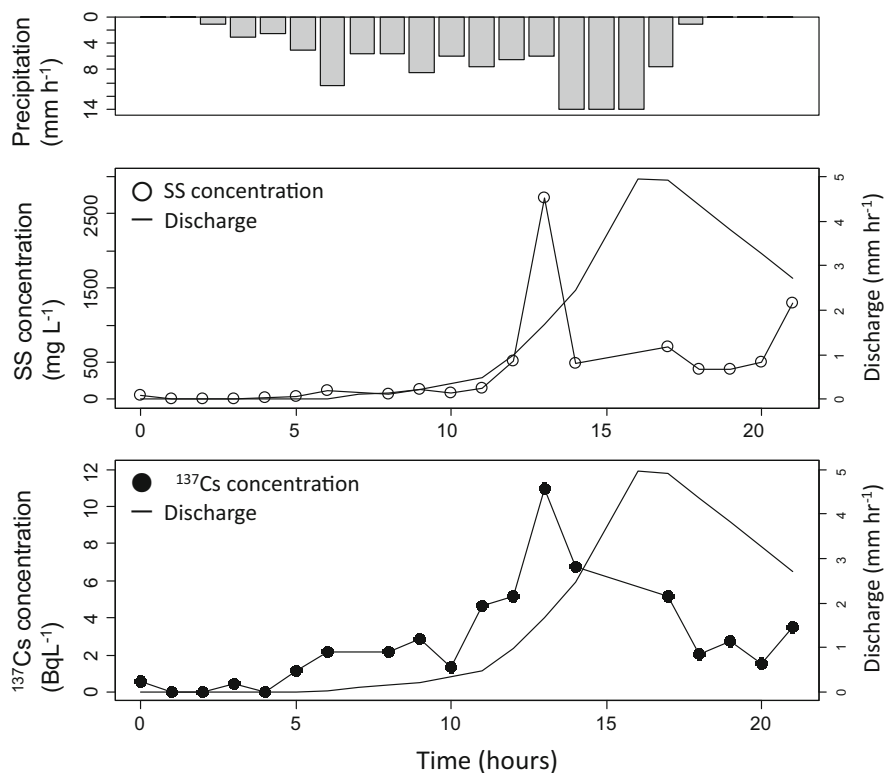


forest floor including the litter layer and the surface layer of mineral soils where the activities of roots and microorganisms are high.

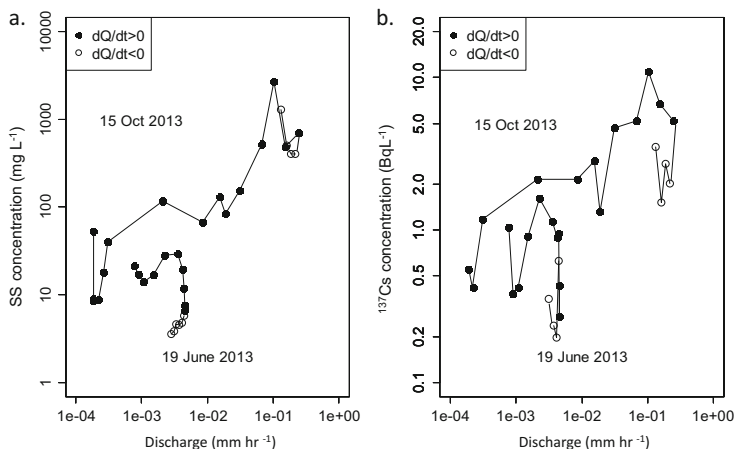
### 14.3.3 <sup>137</sup>Cs Discharge from the Forest Catchment

After the radioactive substances were deposited in March 2011, several reports describing radiocesium transportation through the rivers in Fukushima and surrounding areas were published. One report found that 90 % of the <sup>137</sup>Cs discharged to the coast through the river were attached to suspended solids (SS), and 60 % of those were discharged during flooding (Yamashiki et al. 2014). The reports all stated the importance of evaluating the quantity and quality of SS in order to quantify <sup>137</sup>Cs export from the catchment.

Figure 14.5 represents an example of the short-term temporal change in SS concentration and the <sup>137</sup>Cs concentration with the discharge hydrograph during the storm event on October 15, 2013. The SS concentration increased with an increase



**Fig. 14.5** Temporal variations of precipitation, suspended solid (SS) concentration, <sup>137</sup>Cs concentration, and river discharge rate on October 15, 2013



**Fig. 14.6** Relationships between (1) discharge and suspended solid (SS) concentration, and (2) discharge and  $^{137}\text{Cs}$  concentration. Water samples were collected on June 19 and October 15, 2013.  $dQ/dt > 0$  and  $dQ/dt < 0$  indicate the discharge increasing period and discharge decreasing period, respectively

in river discharge, and began to decrease before the river discharge reached a peak value. The response of  $^{137}\text{Cs}$  concentration to the river discharge was similar to that of the SS concentration, evidencing that the  $^{137}\text{Cs}$  was transported by the SS.

As shown in Fig. 14.6, the changes of the SS and  $^{137}\text{Cs}$  concentrations corresponding to the river discharge rate was different between the period of increasing discharge and decreasing discharge. This suggests that the major source of the discharging  $^{137}\text{Cs}$  was the surface soils and litter detritus from the areas near the riverbank. Those materials can be easily washed out under high flow conditions during a storm.

In the case of the event on October 15, 2013, the decrease in SS and  $^{137}\text{Cs}$  concentrations before the peak discharge moment suggested that the limitation in SS and  $^{137}\text{Cs}$  sources occurred during the increasing discharge period (Figs. 14.5 and 14.6).

The mechanism of washing  $^{137}\text{Cs}$  from the riverbanks might be activated by storm events that occur several to several ten times per year. This means simultaneously that the supply of  $^{137}\text{Cs}$  onto the forest floor of this area (riverbanks) has occurred continuously. The substantial mechanism of this  $^{137}\text{Cs}$  supply was litter fall, throughfall, and stem flow as we explained earlier in this chapter.

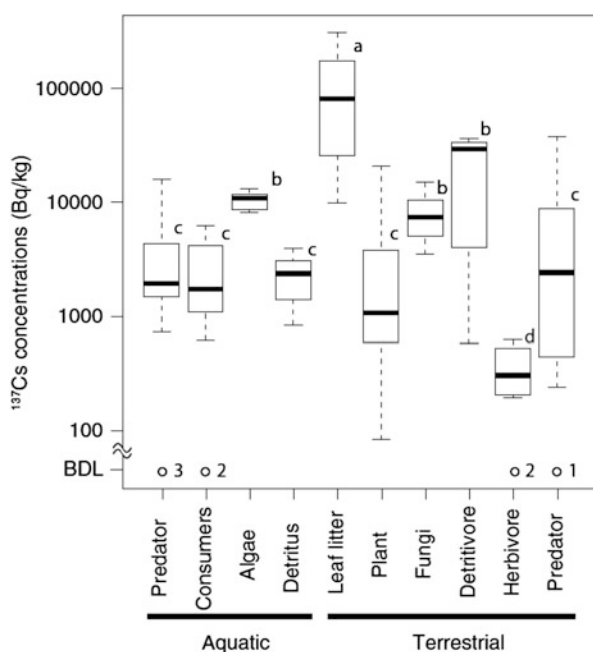
The rating function was established from those relationships between the discharge rate and the  $^{137}\text{Cs}$  concentration. The annual  $^{137}\text{Cs}$  discharge (1-year accumulated value) using this function was estimated to be  $330 \text{ Bq m}^{-2} \text{ year}^{-1}$  for the period from August 31, 2012 to August 30, 2013, while  $670 \text{ Bq m}^{-2} \text{ year}^{-1}$  for the period from October 23, 2012 to October 22, 2013. This difference was attributed to the large storm event in the mid October 2013, which discharged  $227 \text{ Bq m}^{-2}$  of  $^{137}\text{Cs}$  in a single flood event. This emphasizes the importance of quantifying the influence of storm events on the amount of  $^{137}\text{Cs}$  discharged.

The annual  $^{137}\text{Cs}$  discharge through the river was one order of magnitude smaller than the annual total flux of  $^{137}\text{Cs}$  movement from the canopy to the forest floor. This means that the source limitation of the  $^{137}\text{Cs}$  has not occurred for the discharge process on a yearly basis.

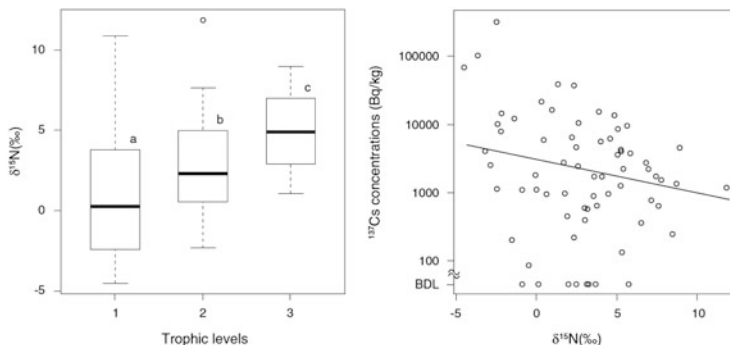
However, the estimated amount of the  $^{137}\text{Cs}$  deposited initially in March 2011 was 100–300  $\text{kBq m}^{-2}$  (Ministry of Education, Culture, Sport, Science and Technology 2013). This is three orders of magnitude larger than the quantity of  $^{137}\text{Cs}$  discharged through the river, indicating that in this forest ecosystem the proportion of the  $^{137}\text{Cs}$  pool that is discharged through the river is significantly less than the proportion lost by radioactive decay.

#### 14.3.4 $^{137}\text{Cs}$ Dispersion Through Food Web

As mentioned above, the  $^{137}\text{Cs}$  has already been in the wood of the dominant trees. It is possible that  $^{137}\text{Cs}$  has begun to circulate among plants and soils. It could be inferred that the food web has already received the  $^{137}\text{Cs}$  dispersion to some extent from the primary producers.



**Fig. 14.7**  $^{137}\text{Cs}$  concentrations in each functional group. Different letters beside each box indicate differences in  $^{137}\text{Cs}$  concentrations based on the grouping of functional groups with the model selection using glm. Data below the detection limit were excluded from the analysis. Numbers beside the BDL (below the detection level) symbols show the number of specimens BDL (Murakami et al. 2014)



**Fig. 14.8** Relationship between  $\delta^{15}\text{N}$  values and  $^{137}\text{Cs}$  concentrations in each sample. Equation:  $\log(^{137}\text{Cs}) = -15.7(\delta^{15}\text{N}) + 1392.3$ . Data below the detection limit were excluded from the analysis (Murakami et al. 2014)

Figure 14.7 shows the observed  $^{137}\text{Cs}$  concentrations in aquatic and terrestrial organisms classified into functional feeding groups. The  $^{137}\text{Cs}$  concentrations in leaf litters, fungi, detritivores, and predators were significantly higher than those in living plant leaves and their herbivores. The large accumulation of  $^{137}\text{Cs}$  in litters and litter detritus indicates that the detrital food chains in the terrestrial community are major pathways that substantially transfer  $^{137}\text{Cs}$ . In the aquatic community, the  $^{137}\text{Cs}$  concentrations in leaf detritus and algae (the base foods) were between those of living leaves and litters, and reflected on those of organisms at higher trophic levels.

As proxies for the relative heights of the trophic levels (Fig. 14.8a), the  $\delta^{15}\text{N}$  values tended to increase as the  $^{137}\text{Cs}$  concentrations in organisms decreased (Fig. 14.8b). This decrease in  $^{137}\text{Cs}$  concentrations through trophic interactions, suggests biological dilution and not accumulation of  $^{137}\text{Cs}$ .

## 14.4 Summary and the Future Directions

Biogeochemical storage, cycling, distribution, and movement of radiocesium in the forested ecosystem were assessed for about 3.5 years after the Fukushima Daiichi Nuclear Power Plant accident. The most important finding was that the  $^{137}\text{Cs}$  movement within the forested ecosystem was still active, and the  $^{137}\text{Cs}$  discharge through the river was significantly small compared to the deposited amount of  $^{137}\text{Cs}$  in this area.

The majority of the  $^{137}\text{Cs}$  pool was in the litter layer and the shallow soil horizons. The attached  $^{137}\text{Cs}$  on the tree canopies and trunks was gradually removed and significantly transferred to the forest floor. Simultaneously,  $^{137}\text{Cs}$  moved into the tree bodies from the surfaces and probably also via root uptake from the litter layer and soils, while  $^{137}\text{Cs}$  had been transported into the new leaves

since the early phase of monitoring. Although the  $^{137}\text{Cs}$  transformation from the tree canopies to the forest floor was still active in 2012 and 2013, it can be inferred that the transferring flux will decrease gradually with the removal of the attached  $^{137}\text{Cs}$ . It can also be hypothesized that the “equilibrium” will be made between the downward  $^{137}\text{Cs}$  transferring flux and the uptake flux by plants. This internal cycling between soils and plants will play important role to retain the relatively mobile forms of  $^{137}\text{Cs}$  in the forest ecosystem, even though certain portion of the  $^{137}\text{Cs}$  in the litter layer moves into the mineral soil horizons, and is fixed strongly by the clay minerals.

Detrital food chains, based on leaf litter detritus as the primary carbon source, have introduced and dispersed  $^{137}\text{Cs}$  through the food web, even to the top predators. However, no evidence of bioaccumulation was detected from the present observations.

An urgent question to address is when and how the  $^{137}\text{Cs}$  diffuses through the grazing food chain and the detritus food chain, and how rapidly does this process occur? The key aims are not only to predict this phenomenon, but also to precisely understand the circulation and redistributions among the system of the soil-plant nutrient cycle, the mechanisms of  $^{137}\text{Cs}$  release from litter and soil organic matter, as well as the  $^{137}\text{Cs}$ -absorbing behavior of plants.

As we reported in this chapter, radiocesium movement within the forest ecosystem is still active and has not reached a steady state. In order to determine the long-term (e.g., equivalent to the  $^{137}\text{Cs}$  half-life) influences of radiocesium existence in the forest ecosystem, we still need to carefully monitor the succession of changes in radiocesium distributions.

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## References

- Bunzl K, Schimmack W, Kreutzer K, Schierl R (1989) Interception and retention of chernobyl-derived  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{106}\text{Ru}$  in a spruce stand. *Sci Total Environ* 78:77–87
- Endo I, Ohte N, Iseda K, Tanoi K, Hirose A, Kobayashi NI, Murakami M, Tokuchi N, Ohashi M (2015) Estimation of radioactive 137-cesium transportation by litterfall, stemflow and throughfall in the forests of Fukushima. *J Environ Radioact* 149:176–185
- Fukuyama T, Takenaka C, Onda Y (2005)  $^{137}\text{Cs}$  loss via soil erosion from a mountainous headwater catchment in central Japan. *Sci Total Environ* 350:238–247

- Hashimoto S, Ugawa S, Nanko K, Shichi K (2012) The total amounts of radioactively contaminated materials in forests in Fukushima, Japan. *Sci Rep* 2:416
- Hashimoto S, Matsuura T, Nanko K, Linkov I, Shaw G, Kaneko S (2013) Predicted spatio-temporal dynamics of radiocesium deposited onto forests following the Fukushima nuclear accident. *Sci Rep* 3:2564
- Hisadome K, Onda Y, Kawamori A, Kato H (2013) Migration of radiocaesium with litterfall in hardwood-Japanese red pine mixed forest and sugi plantation. *J Jpn For Soc* 95:267–274 (In Japanese with English abstract)
- Kato H, Onda Y, Gomi T (2012) Interception of the Fukushima reactor accident-derived  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and  $^{131}\text{I}$  by coniferous forest canopies. *Geophys Res Lett* 39:L20403
- Kinnersley RP, Goddard AJH, Minski MJ, Shaw G (1997) Interception of caesium-contaminated rain by vegetation. *Atmos Environ* 31:1137–1145
- Kitchings T, Digregorio D, Van Voris P (1976) A review of ecological parameters in vertebrate food chains. In: Proceedings of the fourth national symposium on radioecology. Ecological Society of America, Oregon State University, Corvallis, pp 304–313
- Kruyts N, Delvaux B (2002) Soil organic horizons as a major source for radiocesium biorecycling in forest ecosystems. *J Environ Radioact* 58:175–190
- Ministry of Education, Culture, Sport, Science and Technology and Ministry of Agriculture, Forestry and Fisheries (2012) Study report on distribution of radio active substances emitted by the accident of the Fukushima Daiichi Nuclear Power Plant, Tokyo
- Ministry of Education, Culture, Sport, Science and Technology (2013) Distribution map of radiation dose rate. <http://ramap.jmc.or.jp/map/mappdf/pdf/air/20131119/dr/5640-C.pdf>. Accessed 3 Feb 2015
- Murakami M, Ohte N, Suzuki T, Ishii N, Igarashi Y, Tanoi K (2014) Biological proliferation of cesium-137 through the detrital food chain in a forest ecosystem in Japan. *Sci Rep* 4:3599
- Ohte N, Murakami M, Suzuki T, Iseda K, Tanoi K, Ishii N (2012) Diffusion and export dynamics of  $^{137}\text{Cs}$  deposited on the forested area in Fukushima after the nuclear power plant accident in March 2011: preliminary results. In: Proceedings of the international symposium on environmental monitoring and dose estimation of residents after accident of TEPCO's Fukushima Daiichi nuclear power stations. KUR research program for scientific basis of nuclear safety, Kyoto, pp 25–32
- Ohte N, Murakami M, Iseda K, Tanoi K, Ishii N (2013) Diffusion and transportation dynamics of  $^{137}\text{Cs}$  deposited on the forested area in Fukushima after the nuclear power plant accident in March 2011. In: Nakanishi T, Tanoi K (eds) Agricultural implications of the Fukushima nuclear accident. Springer, New York, pp 177–186
- Rowan DJ, Rasmussen JB (1994) Bioaccumulation of radiocesium by fish: the influence of physicochemical factors and trophic structure. *Can J Fish Aquat Sci* 51:2388–2410
- Schimmack W, Förster H, Bunzl K, Kreutzer K (1993) Deposition of radiocesium to the soil by stemflow, throughfall and leaf-fall from beech trees. *Radiat Environ Biophys* 32:137–150
- Soil Survey Staff (2014) Keys to soil taxonomy, 12th edn. USDA-Natural Resources Conservation Service, Washington, DC
- Wakiyama Y, Onda Y, Mizugaki S, Asai H, Hiramatsu S (2010) Soil erosion rates on forested mountain hillslopes estimated using  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{ex}}$ . *Geoderma* 159:39–52
- Wang WX, Ke C, Yu KN, Lam PKS (2000) Modeling radiocesium bioaccumulation in a marine food chain. *Mar Ecol Prog Ser* 208:41–50
- Yamashiki Y, Onda Y, Smith HG, Blake WH, Wakahara T, Igarashi Y, Matsuura Y, Yoshimura K (2014) Initial flux of sediment-associated radiocesium to the ocean from the largest river impacted by Fukushima Daiichi Nuclear Power Plant. *Sci Rep* 4:3714