Diffusion and export dynamics of ^{137}Cs deposited on the forested area in Fukushima after the nuclear power plant accident in March 2011: Preliminary results

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A massive amount of radioactive substances, including cesium-137 (^{137}Cs), emitted from the disabled nuclear power plant, has been deposited on the forested areas in the northeastern region of Honshu Island, Japan after the Fukushima Daiichi nuclear power plant accident. Forests in these regions are particularly important, not only for the forest products industry but also for source areas of drinking water and for residential environments. To clarify the mechanisms of diffusion and export of ^{137}Cs deposited on the forested ecosystem, we initiated intensive field observations in a small catchment that included forest and farmlands. Specifically, we were interested in the Kami-Oguni River catchment that is located in the northern part of Fukushima Prefecture. The following expected major pathways of ^{137}Cs export and diffusion were investigated: 1) transportation of dissolved and particulate or colloidal forms via hydrological processes within a forested catchment and export dynamics through the stream, and 2) diffusion through the food web in terrestrial and aquatic ecosystems of forests. Preliminary findings indicated the following: 1) Most of the ^{137}Cs was discharged as suspended matter. High water flow generated by storm acted to accelerate the transportation of ^{137}Cs from the forested catchments. Thus, the estimation of ^{137}Cs export requires precise evaluation of the high flow acceleration during storm events; 2) Because litter and its detritus may form the biggest pool of ^{137}Cs in the forested ecosystem, ^{137}Cs diffusion occurs more rapidly through the detritus food chain than the grazing food chain. Most predators have already ingested ^{137}Cs, particularly in aquatic environments. An urgent question that needs to be addressed is when and how ^{137}Cs diffuses through grazing food chains and how rapidly this process occurs. To elucidate or to be able to predict these phenomena, the mechanisms of ^{137}Cs release from litter and soil's organic matter need to be clarified.

Keywords: *Fukushima Daiichi nuclear power plant, ^{137}Cs, Forest, Hydrological processes, Food web*

1. Introduction

Following the Fukushima Daiichi nuclear power plant accident that occurred in March 2011, approximately 1.5 × 10^{17} Bq of iodine-131 (^{131}I) and 1.2 × 10^{16} Bq of cesium-137 (^{137}Cs) were emitted into the surrounding environment[1]. These radioactive substances, including ^{137}Cs, were deposited on the forested areas in the northeastern region of Honshu Island, Japan. Forests in these regions are particularly important for humans, not only because of their association with the forestry industry but also because of their importance for residential environment and because these forests are important source areas of drinking water. The first phase of government surveys and investigations showed that the major portion of deposited ^{137}Cs was trapped in the canopy and litter layer at the surface of a soil[1, 2]. ^{137}Cs has been shown to be easily adsorbed onto clay minerals and soil's organic matter[3], which can be transported by eroded soil, particulates and dissolved organic matter through hydrological channels, streams, and rivers[4, 5, 6]. Dissolved ^{137}Cs, which is relatively free from soil adsorption, can also be taken up by microbes, algae, and plants in soil and aquatic systems. By propagating through the food web, this form of ^{137}Cs will eventually be introduced into soil insects and worms, fishes, and birds (Fig. 1).

Namely, it is expected that the major pathways of ^{137}Cs transfer in the forest are following two processes: 1) transportation of via hydrological processes and 2) transfers through the food web in terrestrial and aquatic ecosystems of forests. Dissolved and particulate or colloidal forms of ^{137}Cs are transported by waters within the forest and are partly exported from the forest through hydrological pathways. The food web relationships transfer ^{137}Cs from organisms at the lower trophic level to ones at the higher trophic levels. This also makes ^{137}Cs diffusing from the initial ^{137}Cs pool among organisms.

To precisely describe above mechanisms, field observations of two primary but different pathways were initiated. One studied pathway is the transportation of ^{137}Cs as particulates or dissolved substances via hydrological flow routes. The other pathway is a diffusion of ^{137}Cs through the food web of plants and animals in forested and aquatic ecosystems.

In this paper, we present the concept, the methodology, and the implementation of the field experiment. We also report the results of preliminary investigations that were conducted 15 months after the accident. In our field studies, we focused on investigating the flow of materials through the continuum of forested and stream ecosystems; the materials that we studied included not only ^{137}Cs itself but also some of the related compounds. Currently we are investigating ^{137}Cs transport by water and suspended materials. We also investigate the diffusion of ^{137}Cs through the food webs in the forest–stream ecological continuum.

2. Materials and Methods

(1) Study site

The study was conducted at the Kami-Oguni River catchment in the northern part of Fukushima Prefecture. The area is located approximately 50 km from the Fukushima Daiichi nuclear power plant (Fig. 2a). According to a radioactivity survey report (December 2012) by Ministry of Education, Culture, Sports, Science and Technology that was performed using aircraft survey devices, the air dose rate in this region was 1.9–3.8 μSv h^{-1} and the total deposition rate of ^{137}Cs was 300–600 kBq m^{-2} [7].

The catchment that we studied predominantly comprises forests and paddy fields, and the dominant tree species are broadleaf deciduous trees because the forested regions were mainly used as coppices until the 1960s (Watanabe, personal communication). Some parts of the catchment were used as Japanese cedar and cypress plantations for timber production. The agricultural land also includes orchards, producing peaches and Japanese persimmons.

In 2011, brown rice that is produced in several paddies in this area was found to have high radiocesium concentrations that exceeded the provisional regulation level set by the Ministry of Health, Labour and Welfare of Japan. Based on this survey, Ministry of Agriculture, Forestry and Fisheries decided to prohibit planting the rice plantation during the 2012 season[8]. The Date City government and local farmers only planted test plantations to investigate the rate of ^{137}Cs uptake by rice and the efficacy of countermeasures (such as applying potassium and materials that adsorb ^{137}Cs) in several selected paddies.

The upstream part of the study catchment is mostly forested, whereas the farmland areas, including the paddy fields, are mostly found in the middle to downstream parts of the catchment.

(2) Survey

a) Forested catchment

Within the forested parts of the study catchment, a small subcatchment was selected for intensive observation and sampling (Fig. 2b). Three rectangular plots (20 × 20 m), including two deciduous stands and one cedar plantation, were studied in order to determine the processes involved in ^{137}Cs pools and flows.

To describe the distribution and sizes of the ^{137}Cs pools found within the subcatchment, we periodically collected samples of litter, plants, soil, and stream water and measured the ^{137}Cs concentration in each sample.

To determine fluxes in ^{137}Cs deposition, movement within the subcatchment, and discharge from the catchment, we measured the water flux and ^{137}Cs concentration for every factor that was associated with ^{137}Cs movement, including rain, throughfall, stem flow, and streamwater discharge. The water discharge rate from the subcatchment was continuously measured using a partial flume with a water level gauge. Throughfall and stem flow water were collected from both the secondary deciduous stands and the cedar plantation.

Samples were collected from all members of a terrestrial food web, including litter and living leaves, soil worms, insects, lizards, and snakes.

b) River transect

To describe the riverine distribution of ^{137}Cs concentration, water samples were collected at nine points along the Kami-oguni River (Fig. 2b). Total length of the river transect is 7 km, measured from the outlet of the forest subcatchment. Sampling points were chosen as confluence locations. Samples of aquatic food web members included litter detritus, benthic algae, aquatic macrophytes, benthonic organisms, and fishes.

(3) Analysis

Water samples were filtered using a glass fiber filter (Whatman GF/F, φ = 0.7 μm) immediately after sampling. The ^{137}Cs of suspended matter on the filter was measured. Condensing treatment is required to measure the ^{137}Cs concentration in the dissolved forms using a germanium semiconductor detector. These processes will be conducted in the next phase of the project. Samples of litter and organisms were dried in an oven, then ground and homogenized. Germanium semiconductor detectors were used to measure ^{137}Cs concentration in all samples. Measurements were done at the University of Tokyo and at the National Institute of Radiological Sciences.

For both litter and organisms, we determined the carbon and nitrogen concentrations and the isotope composition (δ^{13}C and δ^{15}N). Isotope data was used to evaluate the trophic levels of sample organisms, and this was done using the methodology described by Lajtha and Michener[9)].

3. Preliminary Results and Discussion

(1) Export of ^{137}Cs by hydrological processes

The observed riverine distribution of ^{137}Cs in suspended matter is shown in Fig. 3. The dataset shown in Fig. 3 includes data collected during a storm event. The median value of each point ranged from 0.4 to 2.6 Bq L^{-1}. The ^{137}Cs concentrations in tributaries from headwaters were higher than those from the main stem. Fig. 4c shows the temporal changes of ^{137}Cs in suspended matter along the Kami-oguni River, during and after a storm event that occurred in June 2012. A large storm event, a typhoon, occurred in the study area from June 19 to June 20 (Figs. 4a and 4b). The increased runoff, caused by this storm event, exported much of the suspended matter from the source areas of the stream, such as the riparian zones of the forested areas (riparian topography is rarely found in agricultural areas because of their well-developed artificial drainage systems). The concentration of ^{137}Cs in the suspended matter increased significantly during the storm event. However, in the middle to downstream parts of the catchment, the concentration was diluted by the water that discharged from the farmlands. The most significant contribution came from the forested catchments that were located in the uppermost regions.

The concentration of ^{137}Cs decreased rapidly over the next day (June 20). This quick response suggests that the mechanism of ^{137}Cs discharge was highly dependent on rapid flows such as the overland flow that occurred in the riparian zones of the forested areas. While this response was rapid, it should be noted that 1 week after the storm event, the concentration of ^{137}Cs in the suspended matter was still higher, as compared to the concentration measured before the storm.

It is generally accepted that, during storm events, the amount of suspended matter increases during conditions of high flow in headwater streams. It is also often observed that the sources of suspended matter are the stream bank and the riparian zone surface. Our data suggest that suspended matter plays an important role in the export of ^{137}Cs from the forested catchment. This interpretation is also supported by a study on other forests in Fukushima, which found that most of the ^{137}Cs pool remained in litter and decomposing organic materials found on the forest floor and soil surface[2].

These findings indicate that high flows generated by storm events accelerate the transportation of ^{137}Cs by water from forested headwater catchments. Precise estimation of the extent of ^{137}Cs export from forested catchments requires a realistic evaluation of high flow acceleration during storm events.

(2) Diffusion of ^{137}Cs through the food web

The δ^{15}N values increased with the expected trophic levels of functional groups of aquatic and terrestrial organisms (Table 1). The ^{137}Cs concentration was significantly higher in terrestrial leaf litter than in the other groups. The concentration of ^{137}Cs in fungi was also high and showed a similar to the one reported previously[1].

The concentration of ^{137}Cs in aquatic organisms (predators and decomposers) was generally high, which clearly indicates that the majority of the aquatic primary consumers feed on litter detritus. On the other hand, the ^{137}Cs concentration in herbivores was low. Those are primary consumers of the terrestrial food web mainly feed on living plant tissues. In other words, the fastest diffusion of ^{137}Cs occurs through the detritus food chain, and it is dominant in the food web of aquatic organisms. In contrast, in terrestrial organisms, herbivorous insects did not ingest ^{137}Cs, although the concentration of ^{137}Cs in terrestrial predators was significantly high. This suggests that many of terrestrial plants have still not yet absorbed ^{137}Cs, and that predators such as lizards and snakes may consume some decomposers found in the shallow soil layers (e.g., angleworms and galley worms that likely consume leaf detritus).

An urgent question that needs to be addressed is when and how ^{137}Cs diffuses through the grazing food chain and how rapidly this process occurs. To predict this phenomenon, mechanisms of ^{137}Cs release from litter and soil's organic matter, as well as the ^{137}Cs absorbing behavior of plants, need to be clarified.

4. Future Studies

Since the Chernobyl nuclear power plant accident, the dynamics of radioactive substances has been monitored in many forested ecosystems in Russia, the Baltic countries, and Scandinavia. Some of these countries have continued long-term ecosystem monitoring[10,11,12]. In southern Finland, for example, it was reported that the Chernobyl fallout remained on site, with more than 3 Bq g^{-1} being found in the uppermost (0–3 cm) humus layer of forest soil[13].

At present, in northeastern Japan, including Fukushima, the largest pool of fallout ^{137}Cs has been found in the litter layer of deciduous forest floors as well as in the canopies of evergreen coniferous forests. Therefore, the urgent and most important objective of future studies will be to predict how fast ^{137}Cs moves from relatively fresh litter to the humus or mineral soils that are found beneath the litter layer. It will also be important to determine how ^{137}Cs is adsorbed to these soils. Although the Scandinavian case studies suggested high retention of fallout in humus, there are geographical differences between Scandinavia and Japan, including differences in climate and geology.

An important point to note is that the total precipitation in Fukushima is twice higher than that in southern Scandinavia, and that ~60% of the total precipitation falls in June, July, August, and September[14,15]. This is a typical seasonal variation caused by the Asian monsoon system. High-temperature summers with high precipitation act to accelerate the decomposition of litter on the forest floor. The transportation of detritus and dissolved organic matter by hydrological pathways is accelerated as well. Thus, in Fukushima, mobilized ^{137}Cs may be more easily transported into the deeper soils and exported by streams, as compared to the transport in Scandinavia.

The geological and geomorphological characteristics of Fukushima forests are also substantially different from those of Finland. Japanese geological settings are generally affected by active orogenic movement. Uplift and erosion activities are much more active than those in Scandinavia and in the northwestern part of the Eurasian continent. This may accelerate the erosion and export of surface materials from the forest floor because of the high precipitation rate in summer[16] .

Thus, due to the environmental differences between the countries, care must be exercised when referencing information that is obtained from Russia, Ukraine, and Scandinavia. For comprehensive comparison between Japan and these countries, intensive and long-term monitoring of various forested ecosystems in the northeastern parts of Japan is required.

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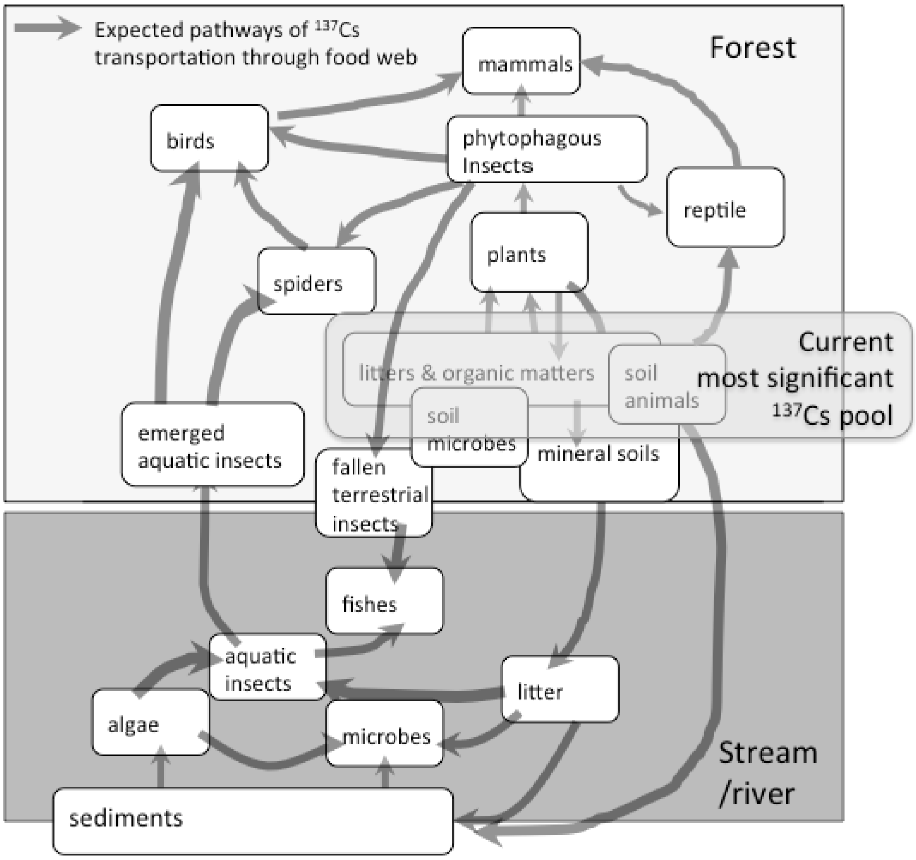

Fig. 1 Conceptual diagram showing the pathways of cesium-137 (^{137}Cs) transfer through the food webs in aquatic and terrestrial ecosystems.

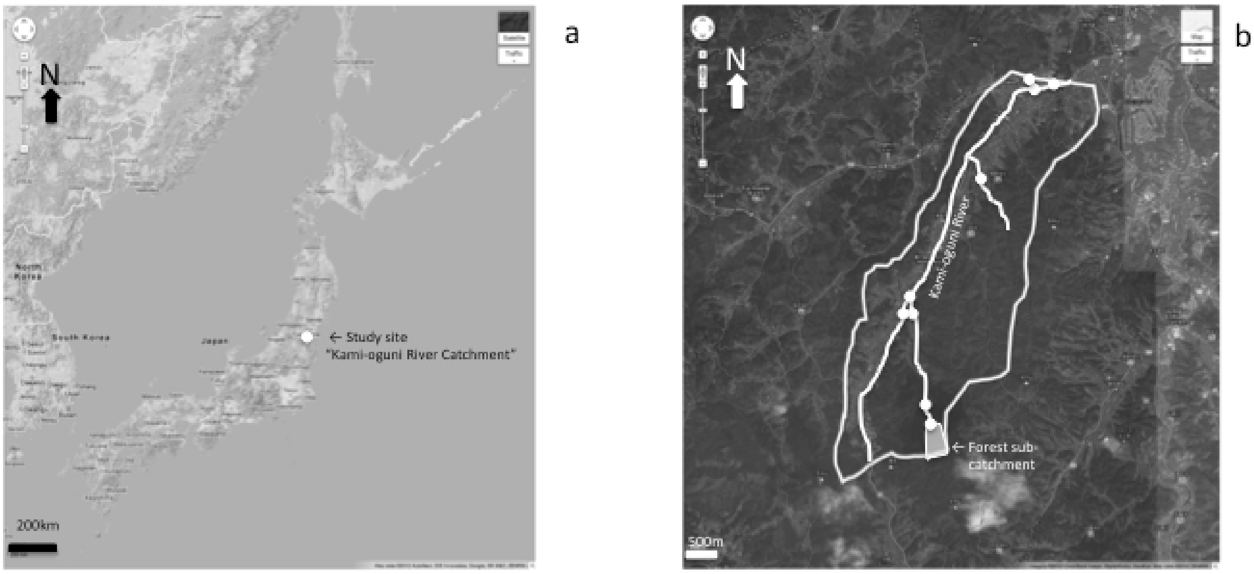

Fig. 2 a) Location of the study site. b) Observation facilities in the study catchment. White dots indicate the water sampling points.

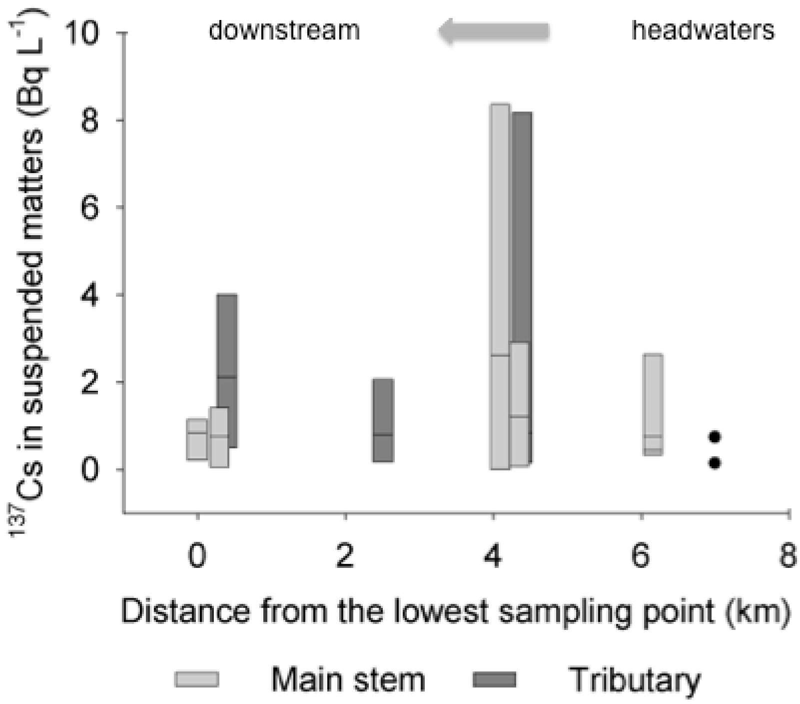

Fig. 3 Boxplot characterization of the observed spatial distribution of cesium-137 (^{137}Cs) concentration in suspended matter along the Kami-oguni River in June 19-July 24, 2012. The boundary of the box closest to zero indicates the 25th percentile, a line within the box marks the median, and the boundary of the box farthest from zero indicates the 75th percentile. The whiskers above and below the box indicate the 90th and 10th percentiles (not shown in these plots).

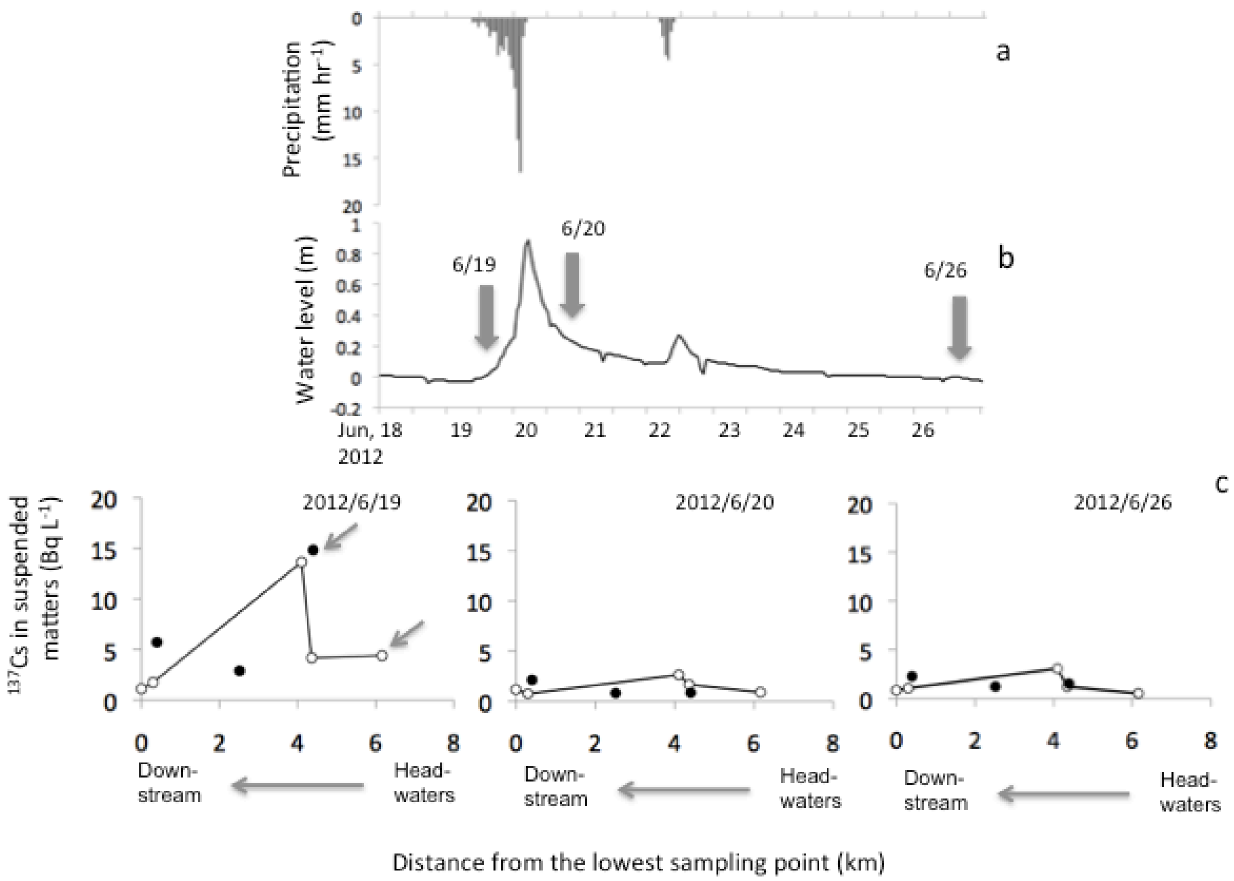

Fig. 4 Changes in spatial distribution of cesium-137 (^{137}Cs) concentration in suspended matter along the Kami-oguni River, during and after a storm event. a) Precipitation at Iidate[15], b) water level of the Hirose River at Ohzeki (downstream of the Kami-Oguni River)[17], c) ^{137}Cs concentration of suspended matter. The arrows on the panel of 2012/6/19 indicate the influxes from the forested headwater catchments.

Table. 1 Nitrogen stable isotope ratio ($\delta^{15}N$) and Cesium-137 (^{137}Cs) concentration of each trophic group of aquatic and terrestrial organisms.

Functional group	$\delta^{15}N$ ($^{o}/_{oo}$)		^{137}Cs concentration (Bq g^{-1})	
	Mean	SD	Mean	SD
		Aquatic		
Leaf litter	-3.2	-	4.07	-
Decomposer	4.0	4.9	1.88	2.34
Predator	5.3	2.1	2.48	3.59
		Terrestrial		
Leaf litter	-3.3	1.0	125.1	134.7
Fungi	2.2	3.9	8.89	5.99
Plant	1.2	3.2	4.22	6.82
Herbivore	0.9	1.6	0.26	0.27
Predator	3.5	2.6	6.03	8.75