



OPEN

SUBJECT AREAS:

FOOD WEBS

FOREST ECOLOGY

ECOSYSTEM ECOLOGY

Received

21 March 2013

Accepted

9 December 2013

Published

8 January 2014

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Biological proliferation of cesium-137 through the detrital food chain in a forest ecosystem in Japan

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Radionuclides, including ¹³⁷Cs, were released from the disabled Fukushima Daiichi Nuclear Power Plant and had been deposited broadly over forested areas of north-eastern Honshu Island, Japan. In the forest, ¹³⁷Cs was highly concentrated on leaf litters deposited in autumn 2010, before the accident. Monitoring of the distribution of ¹³⁷Cs among functional groups clearly showed the role of the detrital food chain as the primary channel of ¹³⁷Cs transfer to consumer organisms. Although many studies have reported the bioaccumulation (or dilution) of radioactive materials through trophic interactions, the present results highlight the importance of examining multiple possible pathways (e.g., grazing vs. detrital chains) in the proliferation of ¹³⁷Cs through food webs. These results provide important insight into the future distribution and transfer of ¹³⁷Cs within forest ecosystems.

A large amount of radionuclide was released from the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident after the disastrous earthquake and subsequent tsunami of March 2011. Among the variety of radionuclides released from FDNPP¹, including iodine, cesium, strontium, and plutonium, cesium 137 (¹³⁷Cs) is the most worrying radionuclide in the environment, with a half-life of 30 years². Unlike iodine, which is mainly found in a gaseous form, cesium rapidly binds to aerosols and is thus washed out by rain from contaminated air masses³ and deposited and accumulated on the land surface⁴. Since most of the Japanese land area is covered by forest, the distribution and transportation of radionuclides within forest ecosystems should be conscientiously monitored (cf. Hashimoto *et al.*⁵ and Ohte *et al.*⁶).

Many previous studies on the distribution and transfer of radionuclides have focused on “bioaccumulation” and the vertical transition between trophic levels^{7–9}. In contrast with this simple view of linear food chains, Polis and Strong¹⁰ incorporated a diverse array of connections among species within complex food webs, which are comprised of grazing and detrital food chains. To explore the biological proliferation of ¹³⁷Cs within forest ecosystems, it is essential to track the ¹³⁷Cs concentrations of each organism component within complex food webs.

In the case of the FDNPP disaster, because the accident occurred in spring (before leaf emergence), the released ¹³⁷Cs would have been deposited on the surface of leaf litters on the forest floor. This accumulated ¹³⁷Cs on leaf litter would likely spread within forest ecosystems through two pathways; grazing and detrital food chains (cf. Polis & Strong¹⁰; Fig. 1). Accumulated ¹³⁷Cs on leaf litter is directly consumed by detritivores and cascades up through detrital food chains. On the other hand, ¹³⁷Cs in an ionic form could be transferred from leaf litter to plants, and thus cascade up through grazing food chains. Therefore, to examine the flow and distribution of radiocesium in a forest ecosystem, the organisms in both terrestrial and aquatic food webs should be carefully monitored. Based on these assumptions, 10 functional groups were chosen as sampling units in the present study (Fig. 1).

In this study, the results of investigations of the Kami-Oguni River catchment in the northern part of the Fukushima Prefecture (Fig. 2) 15 to 18 months after the accident are reported. The main focus of the field observations was the flow of ¹³⁷Cs among organisms in the continuum of forest and stream ecosystems. To understand the distribution and transportation of ¹³⁷Cs within the forest ecosystem, we comprehensively collected the food web components shown in Figure 1 and measured their ¹³⁷Cs radioactivities and nitrogen stable isotope ratio to evaluate trophic transfer among organisms¹¹.

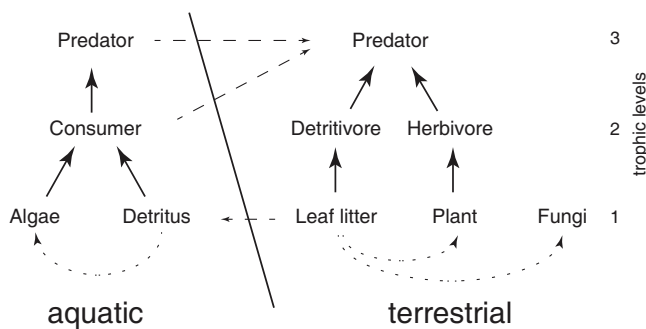


Figure 1 | Schematic food web of the present study. Ten functional groups were used as sampling units. Solid lines indicate trophic interactions and dashed lines indicate nutrient flow. Broken lines show spatial movements; e.g., transformation of tadpoles to frogs and dragonflies or supply of leaf litters from forest to stream.

Results

^{137}Cs was concentrated in the litter layer, especially in the litters of F (fragmented litter) and H (humus) layers, although the highest concentration was observed in the A1 layer at site 3 (Table 1). For the sites in secondary deciduous forests (sites 1 and 3), the highest concentrations were observed in the F and A1 (top horizon of the mineral soil) layers, respectively, but for site 2 in a conifer plantation, the highest concentrations were observed in the F layer, with higher concentrations also observed in the surface L (litter) layer. The total accumulations were 55,100, 25,800 and 28,900 Bq/m² at each site, respectively.

The ^{137}Cs concentrations of each functional group are shown in Figure 3. The glm based on Akaike's Information Criterion (AIC) selected the model with the grouping of functional groups shown in Figure 3 (AIC_{best} = 232.5 vs. AIC_{null} = 265.3). The difference in AIC (ΔAIC) between the second best grouping (terrestrial detritivores as a single independent group with the same grouping as in Figure 3) and the best grouping shown in Figure 3 was 1.9. The ^{137}Cs concentration in leaf litter was highest among all components, reaching more than 300,000 Bq/kg. In the terrestrial food web, ^{137}Cs

concentrations in fungi and detritivores were also high compared with other components. Conversely, the concentration in terrestrial herbivores was lowest among all functional groups. The leaves of living plants also showed relatively low concentrations of ^{137}Cs with high variability among samples. In the stream food web, the ^{137}Cs concentration in algae was high at an average of 10,800 Bq/kg. The concentration in detritus in stream was relatively low, and was much lower than terrestrial leaf litters. The concentrations in stream consumers and predators were also low, at similar levels to detritus in stream.

Nitrogen stable isotope ($\delta^{15}\text{N}$) ratio of the samples was used to confirm trophic levels of sample organisms. Based on this analysis, the glm based on AIC detected the effect of trophic levels on $\delta^{15}\text{N}$ values (AIC_{trophic level} = 378.3 vs. AIC_{null} = 398.1). The $\delta^{15}\text{N}$ did not differ between terrestrial and aquatic food webs since the model segregating terrestrial and aquatic food webs was not selected, with ΔAIC between the habitat + trophic level model (AIC = 378.9) and the trophic level model being 0.6. The relationship between ^{137}Cs concentration and nitrogen stable isotope ratio ($\delta^{15}\text{N}$) is shown in Figure 5. The glm based on AIC detected the correlation between $\delta^{15}\text{N}$ values and ^{137}Cs concentration (AIC _{$\delta^{15}\text{N}$} = 150.3 vs. AIC_{null} = 153.5). In this case, the model segregating the terrestrial and aquatic food webs was not selected, with the AIC of $\delta^{15}\text{N}$ + habitat model being 152.3. The analyses showed that ^{137}Cs decrease with $\delta^{15}\text{N}$ in both terrestrial and aquatic food webs.

Discussion

The present results show that the total accumulation of ^{137}Cs in the studied forest ranged from 26,000 to 55,000 Bq/m². A large heterogeneity in ^{137}Cs concentration was observed within the study area. In the present study, ^{137}Cs was highly accumulated in leaf litters and soils in the F to A1-layers (Table 1). In deciduous forest (sites 1 and 3), these litters corresponded with the leaves deposited in autumn 2010, before the accident. The accident took place on 11 March 2011, immediately before the budbreak of deciduous trees, and the massive release of radionuclides from FDNPP lasted 2 weeks¹². Thus, the ^{137}Cs fell and accumulated on the leaf litters on the forest floor, rather than on the foliage of the forest canopy. This may explain why the ^{137}Cs concentrations in surface leaf litter (A0 to L layers) were less

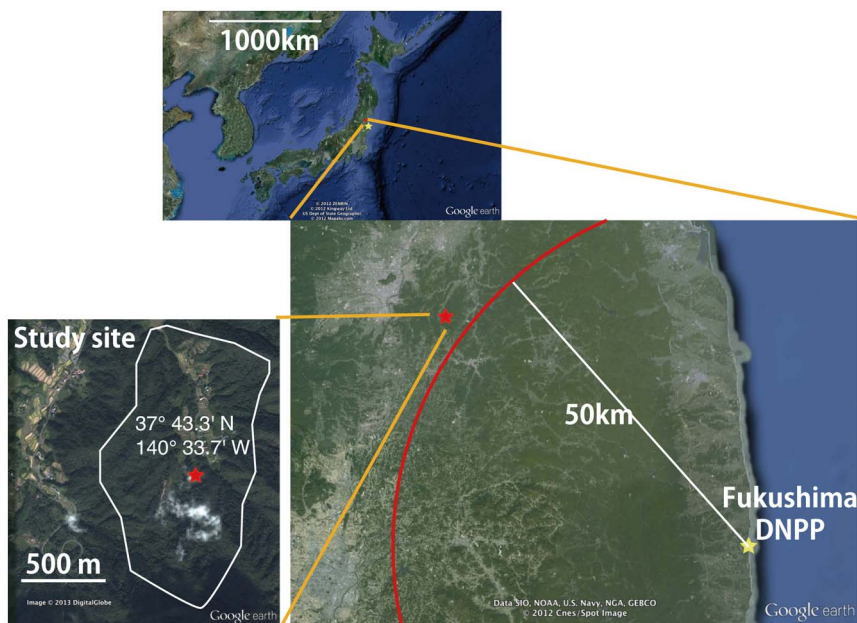


Figure 2 | Maps showing the location of study sites at the Kami-Oguni watershed. The study area was delineated with the geographic coordinates. These maps were attributed to Zenrin, Kingway Ltd., US Dept. of State Geographer, Mapabc.com, DATA SIO, NOAA, U.S. Navy, NGA, GEBCO, Cnes/Spot Image, and DigitalGlobe.



Table 1 | Activities of ^{137}Cs in litter and soil samples. Litter and soil layers were classified following the criterion by Hoover & Lunt³²

Litter and soil layers		Site 1	Site 2	Site 3
		^{137}Cs Bq/m ²		
A0	L	4440	7670	2580
	F	29900	8560	4780
	H	15700	4940	7780
A1		3900	3550	9260
A2		987	1050	4470

than in the lower layers (Table 1). However, in conifer plantation, a higher concentration was observed in the surface L (litter) layer with a lower concentration in the lower layers (F to A2 layers). This may be caused by the interception fraction of the deposited radiocesium by evergreen coniferous canopies¹³.

These contaminated leaf litters on the forest floor were inevitably decomposed by biological activities. According to a review by Aerts¹⁴, the annual decomposition rate of leaf litter in temperate forest is 0.698 ± 0.329 (sd), on average. Applying this value to the present study, about half of ($0.698 \times 0.698 = 0.487$) the leaf litter deposited in autumn 2010 had been decomposed by the autumn of 2012, when the present study finished. Although it is unclear whether biological or geochemical processes contributed to this process, the present results clearly show the input of considerable amounts of ^{137}Cs to the detrital chain through trophic levels (Figs. 1, 3). In terrestrial food webs, highly contaminated leaf litters are consumed by detritivores, e.g. earthworms and Bibionidae fly larvae, which were heavily contaminated with ^{137}Cs . In contrast, the uptake of ^{137}Cs through the grazing chain was limited. Although a large variation in ^{137}Cs concentrations among plant species was observed, the average value of these concentrations was relatively low in plants compared with organisms in the other functional groups (Fig. 3). This was suggestive of a lower uptake of ^{137}Cs from contaminated soils by plants. The herbivores, which consumed the less-contaminated living plant tissues, showed low concentrations of ^{137}Cs . However, predatory animals, such as lizards and snakes, were considerably contaminated with ^{137}Cs . This clearly shows the uptake of ^{137}Cs through the detrital chain, but not through the grazing chain, and the uptake of ^{137}Cs by aboveground generalist predators corresponds with the concept of detrital infusion into grazing food chains¹⁵. Based on studies after the Chernobyl accident, higher contaminations of detritivorous insects than herbivorous and predatory insects were reported^{16–17}. High ^{137}Cs concentrations were also observed in fungi, as has been previously reported^{18–19}. These studies suggested that the higher absorption of potassium by fungi leads to the accumulation of ^{137}Cs in their tissues^{20–21}.

The relationship between ^{137}Cs concentrations and $\delta^{15}\text{N}$ values increases our understanding of the dynamics of radiocesium within the forest ecosystem. The ^{137}Cs concentrations in each sample element decreased with the $\delta^{15}\text{N}$ value (Fig. 5), which showed the increasing trend through trophic levels (Fig. 4). These studies showed the decrease in ^{137}Cs concentrations through trophic interactions, which was suggestive of biological dilution and not accumulation of ^{137}Cs . This supports our conclusion that the detrital food chain is the primary channel of ^{137}Cs transfer to consumer organisms. The relatively high level of ^{137}Cs contamination of predatory organisms in this forest is explained by detrital infusion, but not by the biological accumulation of ^{137}Cs through less contaminated grazing chains.

In the stream food web, algae were highly contaminated with ^{137}Cs , but detritus showed relatively low ^{137}Cs concentrations (Fig. 3), which may have been caused by the relatively low retention and recent accumulation of litter falls into the stream. The retention time of leaf litters within streams is known to be shorter than that on forest

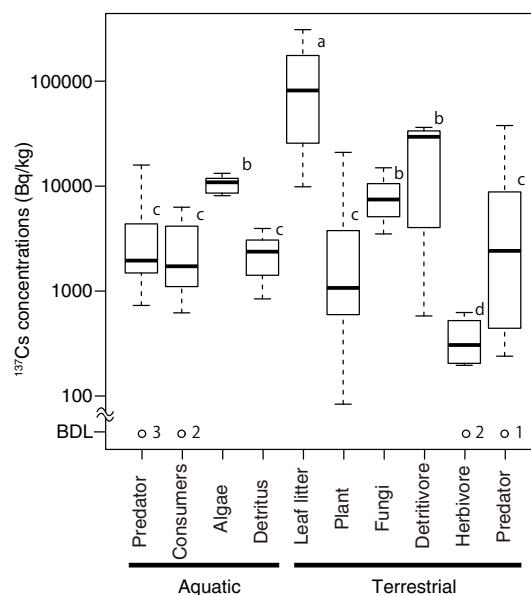


Figure 3 | ^{137}Cs concentrations in each functional group. Different letters beside each box indicate differences in ^{137}Cs concentrations based on the grouping of functional groups with the model selection using glm. The data below the detection limit were excluded from this analysis. Numbers beside the BDL (below detection level) symbols show the number of specimens in BDL.

floors because litter in streams is easily washed out by waterflows²². In contrast, the higher ^{137}Cs concentrations of algae was suggestive of an accumulation of ^{137}Cs . Adam *et al.*²³ reported relatively high accumulation of ^{137}Cs by aquatic algae through the dissolved forms of ^{137}Cs from the contaminated water. Because there were no effective sources of ^{137}Cs within the stream at that moment (Fig. 3), the higher ^{137}Cs concentrations of algae may be caused by leaf litters with higher contamination levels supplied from forest floor. Despite the higher contamination of stream algae, the level of ^{137}Cs concentrations of aquatic consumers and predators were relatively low, which was suggestive of the limited inputs of algal production in stream food webs. The biomass of detritus would be much larger than that of

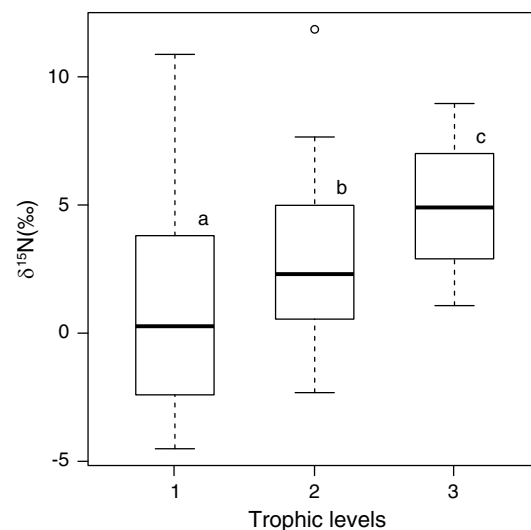


Figure 4 | Changes in $\delta^{15}\text{N}$ value following the trophic levels. The gap in the mean values of trophic levels 1 to 2 was 2.05, and level 2 to 3 was 2.63, respectively. The model selection in glm revealed differences in $\delta^{15}\text{N}$ values among trophic levels.

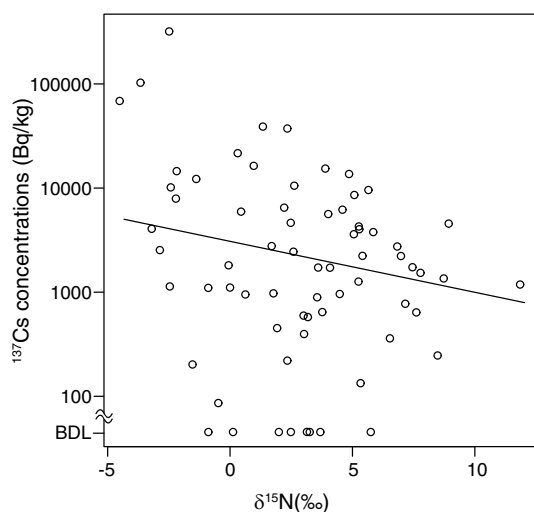


Figure 5 | Relationship between $\delta^{15}\text{N}$ values and ^{137}Cs concentrations in each sample. Equation: $\log(^{137}\text{Cs}) = -15.7(\delta^{15}\text{N}) + 1392.3$. The data below the detection limit were excluded from the analysis.

algae, which caused the relatively low ^{137}Cs contamination in aquatic consumers and predators (Fig. 3).

It should be mentioned that these results are from the very initial processes after ^{137}Cs fallout and in the forest area with an intermediate level of ^{137}Cs contamination. Future studies should examine the intake of ^{137}Cs to the grazing chain and the changes in the degree of transfer of ^{137}Cs through food webs in forest ecosystems. It is important to determine when and how the ^{137}Cs is transferred through grazing food chains. In this study, the ^{137}Cs concentrations of living plants were relatively low (Fig. 3), but those of a fern (*Dryopteris crassirhizoma*) and wild cherry (*Potentilla hebiichigo*) were fairly high compared with the other elements (Table S1). The reason for this variation is unclear, but the depth of the root system²⁴ or mutual interactions with mycorrhizae²⁵ may affect the variation in ^{137}Cs concentrations. The belowground distribution of ^{137}Cs may affect its uptake by plants. Thus, monitoring of the decomposition of the contaminated leaf litters, which are the source of ^{137}Cs , and the subsequent diffusion of ^{137}Cs , are important in predicting the dynamics of ^{137}Cs within forest ecosystems. It is also important to determine whether the transfer rate of ^{137}Cs through the food chain varies over time or among sites with different contamination levels. In the present study, most of the measured food web components were annuals, but several amphibians, reptiles, and other animals and tree species were perennial and may potentially accumulate (or dilute) the ^{137}Cs , as in bioaccumulation⁷. Although many previous studies demonstrated the bioaccumulation of ^{137}Cs by a variety of organisms^{8,9,26} these studies did not examine the effect of food web structure on the distribution of ^{137}Cs among organisms. Furthermore, because the dynamics of ^{137}Cs are known to be affected by the relative amount of the other elements, such as K and stable Cs (^{133}Cs)²⁷, the examination of ^{137}Cs in highly contaminated areas is required to fully understand the distribution and transportation of radionuclides at broader spatio-temporal scales.

Methods

Samples. The study was conducted in a secondary forest at the headwater of the Kami-Oguni River catchment, located 53 km from the FDNPP (Fig. 2). According to a radioactivity survey (December 2012) conducted using aircraft survey devices, the air dose rate in this region was 1.9–3.8 mSv/h and the total deposition rate of ^{137}Cs was estimated as 300,000–600,000 Bq/m²²⁸. The upstream area of the study catchment was composed mostly of forested areas, while farmland consisting mainly of paddy fields was the dominant land used in the middle to downstream area of the catchment. The study area for the samplings was a forest-stream ecotone. In the forest, the dominant tree species are oak (*Quercus serrata*), Japanese zelkova (*Zelkova serrata*), and other broad-leaved deciduous tree species. Some of the forest is used for

plantations of Japanese cedar (*Cryptomeria japonica*) and cypress (*Chamaecyparis obtusa*) for timber production. All samples were taken from the study area and identified to the species and genus level, after which they were classified according to the criteria shown in Fig. 1 (see also Table S1). The sampling points for each sample were selected haphazardly within the study area. The samples were collected from 16 May to 27 July 2012, with the exception of two samples for Bibionidae larvae which were collected on 5 September 2012 and three samples for stream algae which were collected on 27 February 2013.

In addition to these collections, litters and soils of the A0 (L, F and H), A1, and A2 layers, respectively, were sampled from 0.09 m² of forest floor at three sites (sites 1, 2 and 3), which were chosen within the study area; two from secondary deciduous forests (sites 1 and 3) and one from a cedar plantation (site 2). The total amount of ^{137}Cs over a fixed area (1 m²) was calculated from the sum of all layers. The depth of the lower end of the A2 layer was 10 to 12 cm.

All samples were dried for 48 h at 60°C and powdered using a mortar and pestle. For small organisms, several individuals were mixed and powdered together for the measurements. At least 200 mg of samples were gathered and the data on the ^{137}Cs concentrations and nitrogen stable isotope ratio ($\delta^{15}\text{N}$) were obtained from the same sample.

Sample analysis. Germanium semiconductor detectors were used for the measurements of the ^{137}Cs concentrations of all samples. Gamma-ray spectrometry was conducted using germanium detectors (Seiko EG&G). An efficiency calibration of the detectors was made with volume radioactivity standard gamma sources (MX0333U8, Japan Radioisotope Association). The standard reference material JSAC-0471 (the Japan Society for Analytical Chemistry) was used for an accuracy check. The measured values were corrected for the sampling day.

Nitrogen stable isotope ($\delta^{15}\text{N}$) ratios were measured using SerCon ANCA GSL elemental analyser interfaced to a SerCon Hydra 20–20 continuous flow isotope ratio mass spectrometer. Nitrogen isotopic compositions were normalized by using the N-1 standard (=1.36‰) and are reported relative to atmospheric nitrogen²⁹.

Statistical analysis. To determine the best grouping of the functional group in terms of the ^{137}Cs concentration, a generalized linear model (glm) with a normal distribution and log-link function was utilized with the model selection based on Akaike's Information Criterion (AIC). All possible groupings of the combinations of all trophic groups were examined. The model with the lowest AIC value was selected as the preferred grouping³⁰. The effects of trophic levels and the habitat (terrestrial vs. aquatic) on the $\delta^{15}\text{N}$ values were analysed using glm. The best model explaining the variation of $\delta^{15}\text{N}$ values was selected based on AIC. The relationship of $\delta^{15}\text{N}$ value and ^{137}Cs concentrations with examining the effect of the habitat (terrestrial vs. aquatic) were also analysed using glm. The best model explaining the variation of ^{137}Cs concentrations was selected based on AIC. All analyses were performed in R version 2.13.1³¹.

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Acknowledgments

This study was supported by a grant (24248027) for scientific research from the Ministry of Education, Culture, Sports, Science and Technology Japan Society for the Promotion of Science.

Author contributions

M.M. and N.O. designed the study and wrote the manuscript with input from all other authors. T.S. and Y.I. collected samples and performed the calculations. N.I. and K.T. conducted the radioisotope analyses.

Additional information

Supplementary information accompanies this paper at <http://www.nature.com/scientificreports>

Competing financial interests: The authors declare no competing financial interests.

How to cite this article: Murakami, M. *et al.* Biological proliferation of cesium-137 through the detrital food chain in a forest ecosystem in Japan. *Sci. Rep.* **4**, 3599; DOI:10.1038/srep03599 (2014).



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