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

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Radionuclides, including \textsuperscript{137}Cs, 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, \textsuperscript{137}Cs was highly concentrated on leaf litters deposited in autumn 2010, before the accident. Monitoring of the distribution of \textsuperscript{137}Cs among functional groups clearly showed the role of the detrital food chain as the primary channel of \textsuperscript{137}Cs 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 \textsuperscript{137}Cs through food webs. These results provide important insight into the future distribution and transfer of \textsuperscript{137}Cs 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 FDNPP\textsuperscript{1}, including iodine, cesium, strontium, and plutonium, cesium 137 (\textsuperscript{137}Cs) is the most worrying radionuclide in the environment, with a half-life of 30 years\textsuperscript{2}. 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 masses\textsuperscript{3} and deposited and accumulated on the land surface\textsuperscript{4}. 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.\textsuperscript{5} and Ohte et al.\textsuperscript{6}).

Many previous studies on the distribution and transfer of radionuclides have focused on "bioaccumulation" and the vertical transition between trophic levels\textsuperscript{7–9}. In contrast with this simple view of linear food chains, Polis and Strong\textsuperscript{10} 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 \textsuperscript{137}Cs within forest ecosystems, it is essential to track the \textsuperscript{137}Cs 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 \textsuperscript{137}Cs would have been deposited on the surface of leaf litters on the forest floor. This accumulated \textsuperscript{137}Cs on leaf litter would likely spread within forest ecosystems through two pathways: grazing and detrital food chains (cf. Polis & Strong\textsuperscript{10}; Fig. 1). Accumulated \textsuperscript{137}Cs on leaf litter is directly consumed by detritivores and cascades up through detrital food chains. On the other hand, \textsuperscript{137}Cs 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 \textsuperscript{137}Cs among organisms in the continuum of forest and stream ecosystems. To understand the distribution and transportation of \textsuperscript{137}Cs within the forest ecosystem, we comprehensively collected the food web components shown in Figure 1 and measured their \textsuperscript{137}Cs radioactivities and nitrogen stable isotope ratio to evaluate trophic transfer among organisms\textsuperscript{11}.
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/m$^2$ 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 = 378.5 vs. AIC null = 398.1). 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 ($\delta^{15}$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 $\delta^{15}$N values (AIC trophic level = 378.3 vs. AIC null = 398.1). The $\delta^{15}$N did not differ between terrestrial and aquatic food webs since the model segregating terrestrial and aquatic food webs was not selected, with $\Delta$AIC 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 ($\delta^{15}$N) is shown in Figure 5. The glm based on AIC detected the correlation between $\delta^{15}$N values and $^{137}$Cs concentration (AIC $\delta^{15}$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 $\delta^{15}$N + habitat model being 152.3. The analyses showed that $^{137}$Cs decrease with $\delta^{15}$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/m$^2$. 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 weeks. 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

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**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.
Aerts14, the annual decomposition rate of leaf litter in temperate forests is known to be shorter than that on forest floors because litter in streams is easily washed out by waterfows22. In contrast, the higher 137Cs concentrations of algae was suggestive of an accumulation of 137Cs. Adam et al.23 reported relatively high accumulation of 137Cs by aquatic algae through the dissolved forms of 137Cs from the contaminated water. Because there were no effective sources of 137Cs within the stream at that moment (Fig. 3), the higher 137Cs 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 137Cs 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 detritivorous insects than herbivorous and predatory insects were reported16–17. High 137Cs concentrations were also observed in fungi, as has been previously reported18–19. These studies suggested that the higher absorption of potassium by fungi leads to the accumulation of 137Cs in their tissues10–21.

The relationship between 137Cs concentrations and δ15N values increases our understanding of the dynamics of radioesium within the forest ecosystem. The 137Cs concentrations in each sample element decreased with the δ15N value (Fig. 5), which showed the increasing trend through trophic levels (Fig. 4). These studies showed the decrease in 137Cs concentrations through trophic interactions, which was suggestive of biological dilution and not accumulation of 137Cs. This clearly shows that detrital food chain is the primary channel of 137Cs transfer to consumer organisms. The relatively high level of 137Cs contamination of predatory organisms in this forest is explained by detrital infusion, but not by the biological accumulation of 137Cs through less contaminated grazing chains.

In the stream food web, algae were highly contaminated with 137Cs, but detritus showed relatively low 137Cs 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 floors because litter in streams is easily washed out by waterfows22. In contrast, the higher 137Cs concentrations of algae was suggestive of an accumulation of 137Cs. Adam et al.23 reported relatively high accumulation of 137Cs by aquatic algae through the dissolved forms of 137Cs from the contaminated water. Because there were no effective sources of 137Cs within the stream at that moment (Fig. 3), the higher 137Cs 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 137Cs 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 detritivorous insects than herbivorous and predatory insects were reported16–17. High 137Cs concentrations were also observed in fungi, as has been previously reported18–19. These studies suggested that the higher absorption of potassium by fungi leads to the accumulation of 137Cs in their tissues10–21.

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Table 1 | Activities of 137Cs in litter and soil samples. Litter and soil layers were classified following the criterion by Hoover & Lunt.32

| Litter and soil layers | Site 1 | Site 2 | Site 3 |
|-----------------------|--------|--------|--------|
| A0                    | L      | 4440   | 7670   | 2580   |
|                       | F      | 29900  | 8560   | 4780   |
|                       | H      | 15700  | 4940   | 7780   |
| A1                    |       | 3900   | 3550   | 9260   |
| A2                    |       | 987    | 1050   | 4470   |

Figure 3 | 137Cs concentrations in each functional group. Different letters beside each box indicate differences in 137Cs 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.

Figure 4 | Changes in δ15N 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 δ15N values among trophic levels.
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 ($D. crassirhizoma$) and wild cherry ($D. micrantha$) were also analyzed using glm. The best model explaining the variation of $^{137}$Cs concentrations was selected based on AIC. All analyses were performed in R version 5.1.3.

**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/m². The upstream area of the study catchment was composed mostly of forested areas, while farmlands consisting mainly of paddy fields was the dominant land used in the middle to downstream area of the catchment. The sample area for the samplings was a forest-stream ecotope. In the forest, the dominant tree species are oak ($Q. serrata$), Japanese zelkova ($Z. serrata$), and other broad-leaved deciduous tree species. Some of the forest is used for plantations of Japanese cedar ($C. japonica$) and cypress ($C. obtusa$) 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 was gathered and the data on the $^{137}$Cs concentrations and nitrogen stable isotope ratio ($\delta^{15}$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 (Sego EG&G). An efficiency calibration of the detectors was made with a volume radioactivity standard gamma sources (MX0333US, 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 ($\delta^{15}$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 nitrogen.

**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 grouping. The effects of trophic levels and the habitat (terrestrial vs. aquatic) on the $\delta^{15}$N values were analysed using glm. The best model explaining the variation of $\delta^{15}$N values was selected based on AIC. The relationship of $\delta^{15}$N values 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 2.13.1.
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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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