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Estimation of spatio-temporal distribution of ^{137}Cs concentrations in litter layer of forest ecosystems in Fukushima using FoRothCs model ☆

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ABSTRACT

The nuclear power plant accident in Fukushima had led to pollution of forest ecosystems with ^{137}Cs in 2011. In this study, we simulated the spatiotemporal distribution of ^{137}Cs concentrations of litter layer in the contaminated forest ecosystems in two decades from 2011, which is one of the key environmental components of ^{137}Cs migration in the environment due to the high bioavailability of ^{137}Cs in the litter. Our simulations showed that ^{137}Cs deposition is the most important factor in the degree of contamination of the litter layer but vegetation type (evergreen coniferous/deciduous broadleaf) and mean annual temperature are also important for changes over time. Deciduous broadleaf trees had higher initial concentrations in the litter layer due to the direct initial deposition on the forest floor. However, the concentrations remained higher than those in evergreen conifers after 10 years due to redistribution of ^{137}Cs by vegetation. Moreover, areas with lower average annual temperatures and lower litter decomposition activity retained higher ^{137}Cs concentrations in the litter layer. The results of the spatiotemporal distribution estimation of the radioecological model suggest that, in addition to ^{137}Cs deposition, elevation and vegetation distribution should also be considered in the long-term management of contaminated watersheds, which can be informative in identifying hotspots of ^{137}Cs contamination on a long-term scale.

1. Introduction

As a result of the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, radioactive fallout had spread on land, in which 70% of the contaminated area consisted of forest ecosystems (Hashimoto et al., 2012). After the FDNPP accident, most deposited ^{137}Cs in the forest ecosystem moved to the forest floor within a few years due to physical and biological weathering (Onda et al., 2020). Although some ^{137}Cs were discharged into rivers as a result of water movement, compared to the ^{137}Cs deposited in the catchment area, the percentage of ^{137}Cs discharged and lost from the system was < 0.3% per year, and most ^{137}Cs remained in the forest ecosystem (Onda et al., 2020). The physical half-life of ^{137}Cs is approximately 30.1 years, and it contaminates the environment in the long-term when released into the forest ecosystem (Hashimoto et al., 2020c). Therefore, long-term monitoring is important to understand the contamination of ecosystems because ^{137}Cs is cycled and redistributed within the forest ecosystem.

In forest ecosystems affected by the Chernobyl accident, large amounts of ^{137}Cs have been found to persist in the litter layer (i.e., organic layer) of the forest floor for several decades, and the litter layer is considered to be a major reservoir of the ^{137}Cs inventory in the forest (Rafferty et al., 2000; Goor and Thiry, 2004; Karadeniz et al., 2015). Conversely, it has been noted that, in forests contaminated by radioactive materials due to the FNDPP accident, the rate of migration from the organic layer to mineral soil may generally be faster than that at the Chernobyl site (Koarashi et al., 2016; Konoplev et al., 2016; Coppin et al., 2016; Takahashi et al., 2018; Yoschenko et al., 2018; Koarashi and Atarashi-Andoh, 2019; Imamura et al., 2020; Manaka et al., 2022). However, although various environmental factors are contributing to the accumulation of ^{137}Cs in the organic layer, their effects have not

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been quantitatively determined from a long-term perspective (Shcheglov, 1999; Imamura et al., 2020; Manaka et al., 2022). From recent studies, the reduction rate in ^{137}Cs inventory in the litter layer also varies greatly depending on the environment (Koarashi et al., 2016; Takahashi et al., 2018; Imamura et al., 2020; Manaka et al., 2022). For example, Ito et al. (2018) pointed out that ^{137}Cs accumulation is greater at the thicker litter layer, indicating that the decomposition characteristics of the litter is a key factor in the ^{137}Cs dynamics in the litter layer.

Regardless of the amount of ^{137}Cs inventory, it is quite important to understand the concentration of ^{137}Cs in the litter in order to consider the ^{137}Cs dynamics and effects in the ecosystem. It should be noted that the organic materials in the litter layer keep the high radioactivity concentration per unit weight of any ecosystem components in the forest ecosystem (Hashimoto et al., 2020c), which shows the high bioavailability. For example, the contaminated litter layer in forest ecosystems has a significant contribution on the ^{137}Cs concentration in detritus aquatic insects in riverine ecosystems (Ishii et al., 2017; Kurikami et al., 2019; Sakai et al., 2021). Moreover, submerged litter in the riparian zone can leach the dissolved forms of ^{137}Cs to the downstream water (Sakai et al., 2016; Tsuji et al., 2016). They are acting as a secondary source of dissolved ^{137}Cs in the river. Thus, the litter is regulating both the ^{137}Cs concentrations in the particle matters and dissolved ^{137}Cs concentrations in the downstream river ecosystem of the contaminated forest (Sakai et al., 2016; Tsuji et al., 2016; Gomi et al., 2018; Sakai et al., 2021; Hayashi et al., 2022). Therefore, it is important to understand the time evolution of the ^{137}Cs concentration in the litter layer to consider environmental dynamics and ecological effects.

Although there are already several radioecological models that can predict the ^{137}Cs concentration in the organic layer (litter layer) of the forest floor, including some studies in forests affected by the Chernobyl nuclear power plant accident (IAEA, 2002; Hashimoto et al., 2021; Ota and Koarashi, 2022), the ^{137}Cs concentration in the litter layer has not yet been adequately verified spatially in the area contaminated by the FNDPP. Currently, more than 10 years after the FNDPP accident, the number of observations on the concentrations of ^{137}Cs in forest ecosystems contaminated by the FNDPP accident has accumulated, which enables to verify the results of the spatial estimates in this region (Hashimoto et al., 2020b). In this study, we tried to estimate the spatiotemporal ^{137}Cs concentration in the litter layer in the contaminated area in Fukushima Prefecture using the radio-ecology model "FoRothCs" (Nishina and Hayashi, 2015). The model couples biomass/carbon dynamics with ^{137}Cs dynamics, which allows estimation of the concentration in the litter layer in forest ecosystems (Nishina and Hayashi, 2015). In our previous studies (Nishina et al., 2018; Hashimoto et al., 2021), FoRothCs has been calibrated by observed ^{137}Cs concentrations in leaves, branches, trunks, litter, and litter layers of conifers and deciduous broadleaf forests and in soil in the monitoring campaign by the Forestry Agency (Imamura et al., 2017). Using ^{137}Cs deposition data modified by Kato and Onda (2018) (Fig. 2), this study evaluated the decadal evolution of ^{137}Cs concentrations in litter in two vegetation types, evergreen coniferous and deciduous broadleaf trees, for areas with ^{137}Cs deposition exceeding 10 kBq m^{-2}, over the decade following the accident. From the simulation, we aimed to quantitatively understand the contamination status of litter in the FNDPP accident.

2. Material and methods

2.1. Model

In this study, we applied the FoRothCs model (Nishina and Hayashi, 2015; Nishina et al., 2018; Hashimoto et al., 2021) to estimate the ^{137}Cs concentrations in the litter layer in forest ecosystems in Fukushima Prefecture. The FoRothCs model consists of seven compartments (leaves, branch, stem, litter, soil (organic, mineral soils), and microbes), and these compartments are interconnected by the ^{137}Cs migration processes. This model has biomass growth scheme and litter (and soil) carbon decomposition schemes, which govern the ^{137}Cs dynamics in the litter layer. The time step of this model is monthly and requires monthly air temperature and precipitation.

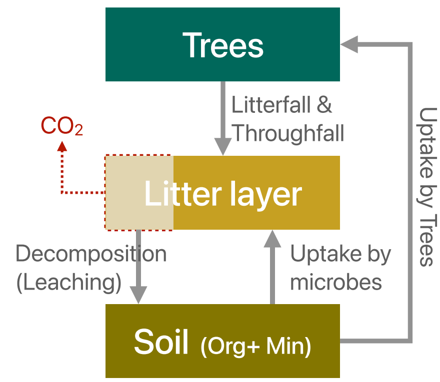

Fig. 1. Schematic diagram of migration process for litter ^{137}Cs dynamics in FoRothCs model.

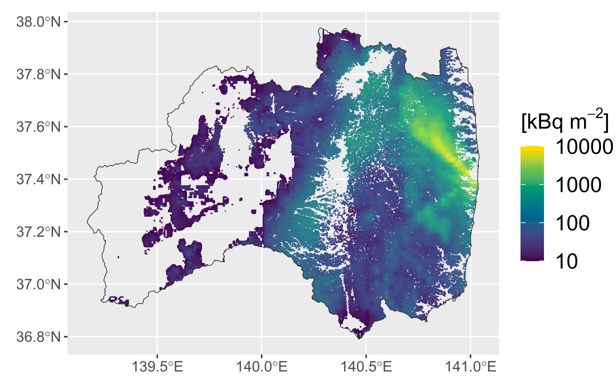

Fig. 2. Initial ^{137}Cs deposition (over 10 kBq m^{-2}) at 2011 in forested areas in Fukushima Prefecture estimated by Kato and Onda (2018). The color gradient indicates the amount of deposition.

The migration process of the FoRothCs model regarding ^{137}Cs dynamics in the litter is shown in Fig. 1 as a schematic diagram. ^{137}Cs inputs to the litter layer, apart from initial deposition, are mainly litterfall from the canopy and upward migration by microbial activity from the soil in this model. The model does not explicitly address throughfall associated with the movement of water from the litter layer to the soil layer (Kurihara et al., 2018b) but assumes that it is included in what moves with decomposition. More detailed model information, including specific functions, can be found in two previous references (Nishina and Hayashi, 2015; Nishina et al., 2018).

2.2. Input data

The simulation domain is according to the spatial initial ^{137}Cs deposition dataset developed by Kato and Onda (2018), which covers the contaminated forest area with ^{137}Cs deposition > 10 kBq m^{-2} in Fukushima Prefecture with 250 m × 250 m mesh. This initial deposition was reconstructed by the two different timing surveys of the third (July 2, 2011) and fifth (June 28, 2012) airborne monitoring for ^{137}Cs deposition density with correction of the decay rates. In the contaminated area of the FNDPP accident, planted forests occupy 50% of the forest area, with cedar forests, red pine forests, and cypress forests as the main vegetation types. These conifers are evergreen conifer trees. In natural forests (mostly secondary forests) in Fukushima, deciduous broadleaf trees, such as *Quercus serrata* are dominant. In the simulation, we divided the vegetation types to coniferous and deciduous trees according to the vegetation map in Hashimoto et al. (2020a). We assumed 70% of initial deposition was intercepted by the canopy (leaves and branch) in evergreen conifer forest according to the scenario in Gonze et al. (2021); Hashimoto et al. (2021). Conversely, for deciduous broadleaf forest, we assumed 10% of the initial deposition to be intercepted by the branch, because the leaves were not expanded at the time of deposition.

The maximum tree height was estimated by the site index, which is estimated by the empirical equations with geographical variables, such as elevation, topography, soil, and other location and environmental factors that greatly affect the productivity of trees. For the site index score (i.e., coefficients of environmental variables in the linear model for tree heights) table of evergreen conifer, we used the score table of Japanese cedar forests in Niigata Prefecture (Ito, 2018), which is adjacent to Fukushima Prefecture, and the score table of oak trees in Iwate Prefecture for deciduous brodleaf trees (Todate et al., 1986). The scores used in this study are summarized in Table S1 for evergreen conifer forest and Table S2 for deciduous broadleaf forest. For elevation, slope-angle, slope direction, and soil type, we used open data in the digital national land information provided by the Ministry of Land, Infrastructure, Transportation and Tourism of Japan (MLIT, 2022). The data spatial resolution is 250 × 250 m. The elevation data was obtained by resampling a 10 m by 10 m spatial resolution mesh to a 250 m resolution mesh, and the ground slope data was obtained by the same process. Forest age was obtained from forest plan maps for national forests (MLIT, 2022) and forest registers for private forests. These are shape file format and converted to mesh as input data. We set the different decomposable plant material (DPM) and resistant plant material (RPM) ratio in the litterfall of each conifer and deciduous tree according to Shirato and Yokozawa (2006). We used 64 for conifer and 49 for deciduous trees (originally in *Quercus mongolica*) in the DPM:RPM ratio. For climate variables, we used the Mesh Climate Value 2010 for monthly mean air temperature and monthly precipitation in the MLIT database (MLIT, 2022), which contains 30-year averages from 1981 to 2010 in 1-km^2 resolution mesh grid data.

2.3. Spin-up and simulation

In previous studies, parameters for the radiocesium dynamics of FoRothCs have been determined using approximate Bayesian calibration for evergreen conifers and deciduous broadleaf trees, respectively (see Nishina et al. (2018) for methodology). In this study, the parameters used in the model intercomparison study (Hashimoto et al., 2021) were retained, using mean values from the meta-analysis provided by Gonze et al. (2021) for evergreen conifers and observed values of Konara oak (*Quercus serrata*) from Imamura et al. (2017) for deciduous broadleaf trees. The parameters regarding the ^{137}Cs migrations were calibrated using Bayesian approximate computation. The parameter values used in this study are summarized in Table S3. In addition, Fig. S1 shows a comparison between simulations with these parameters and observed data regarding the ^{137}Cs concentration and litter mass of the litter layer.

The initial litter layer and soil carbon were estimated by 200 years spin-up in FoRothCs. The initial values was set when the forest has grown sufficiently and soil and litter carbon has reached almost equilibrium. For the spin-up period, we used the same climate data repeatedly. After a spin-up simulation, we run FoRothCs for the year of forest age in each grid (provided by the input data) before the FNDPP accident in March 2011. After this procedure, we run FoRothCs for 20 years. In this study, once all forested areas were calculated for each vegetation type, these 2 maps were harmonized according to the vegetation map (Hashimoto et al., 2020a) after the calculation to obtain the final results.

2.4. Validation

In this study, the model was validated using a database “CsDB” of publicly available observation data for the ^{137}Cs concentrations of the litter layer from 2011 to 2017 after the Fukushima accident, collected by Hashimoto et al. (2020b). Additionally, 84 unpublished records analyzed by the authors, including data from 2012 to 2020, were used in the analysis. These are provided as a csv file in the supplement material. Model outputs were compared with observations matched for geographic location, vegetation, and dates of observations. It should be noted that the detailed conditions of the observation forest sites (e.g., forest age, tree density, slope-angle, slope direction) in the database were not reflected in the simulations. Since these conditions are not available for all sites, they were not taken into account in this evaluation.

We calculated correlation coefficients ρ and root mean squared logarithmic error (RMSLE) to check the model performance in litter ^{137}Cs concentrations in each tree type. RMSLE is defined as follows:

$$RMSLE = \sqrt{\frac{1}{N}\sum_{n=1}^{N}\left(log(Y_{pred}+1) - log(Y_{obs}+1)\right)^2},$$

where Y_{pred} and Y_{obs} are the simulated and observed ^{137}Cs concentrations, respectively and N is the sample size.

3. Result and discussion

3.1. Spatial-temporal ^{137}Cs distribution of the litter layer

The estimated ^{137}Cs concentrations were generally agreed with the observed values in the CsDB database (Hashimoto et al., 2020a), in which the correlation coefficients (rho) were significant at p<0.05 in both vegetation types. The RMSLEs were 0.856 for evergreen conifer and 1.036 for deciduous broadleaf trees, respectively (Fig. 3). The spatial mesh data used in this simulation is based on publicly registered information for each forest stands (e.g., age, density, etc.), while the CsDB database contains more detailed and accurate information for each forest stand based on field surveys in some records. It should be noted that the forest stand information in CsDB does not exactly match the spatial mesh data used in this study. Despite this, the simulated ^{137}Cs concentrations were generally agreed with observations for both vegetation types. However, in the deciduous broadleaf forests in 2016, there are significant discrepancies between some observations in the CsDB and the simulated values, which are significantly overestimated by the model. The observations considerably overestimated by FoRothCs in 2016 are originally provided by the Forestry Agency, which report includes 23 observations for litter ^{137}Cs concentrations from the same site, with ranges from 0.1 kBq kg^{-1} to 3.9 × 10 kBq kg^{-1} (tree species is *Cerasus jamasakura*). There are many potential factors that could generate such large variations at an observation site, for example, microtopography would be a possible factor (Koarashi et al., 2014). However, at the spatial resolution of the simulations in this study, the same mesh is calculated under all the same conditions. On the other hand, there are inherent uncertainties in the observations. It is possible that some of the observed values are not representative of the litter layer at the observation site, e.g. the litter sample was at a very early stage of decomposition.

The simulated ^{137}Cs concentrations in the litter layer of contaminated evergreen coniferous forest varied from 4.22×10^{-1} kBq kg^{-1} to 7.16×10^{3} kBq kg^{-1} in March 2011. Conversely, those of deciduous broadleaf forest in the same period ranged from 9.37×10^{-1} kBq kg^{-1} to 1.82×10^{4} kBq kg^{-1}. The median (mean) of ^{137}Cs concentrations in the litter layer in 2011 were 1.31 × 10 (5.13 × 10) kBq kg^{-1} in the evergreen coniferous forest and 3.33 × 10 (1.27×10^{2}) kBq kg^{-1} in the deciduous broadleaf forests, respectively. Thus, the median and mean initial litter ^{137}Cs concentrations were higher in deciduous broadleaf

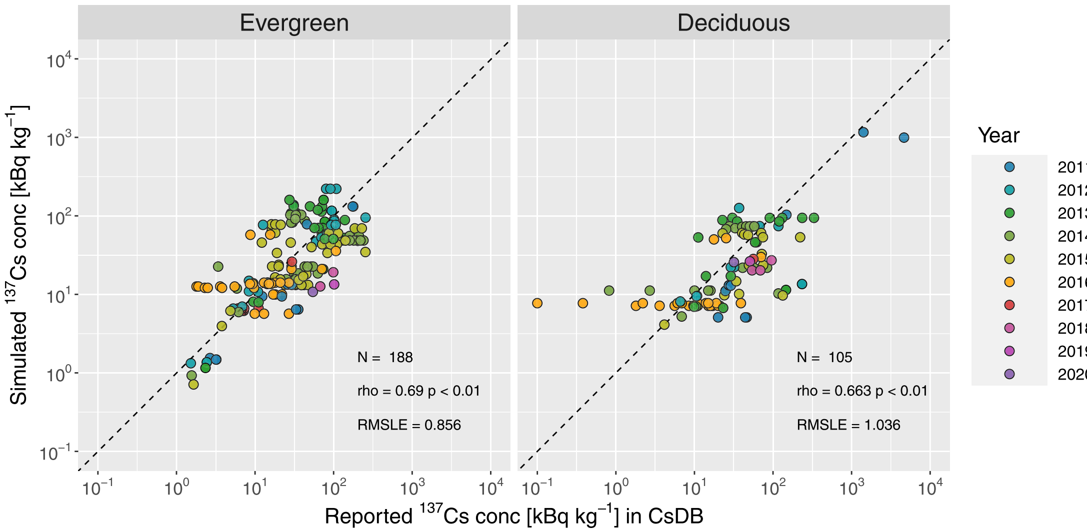

Fig. 3. Comparison between observed and simulated litter ^{137}Cs concentrations in each vegetation type.

forests, mainly due to the fact that deciduous broadleaf trees were in the pre-leafing stage at the time of the accident and ^{137}Cs deposition were not captured in the forest canopy during the deposition events in March, 2011. For the evergreen conifer forests in this study, 70% of the initial deposition is assumed to be trapped once in the forest canopy according to Gonze et al. (2021). The spatial patterns of both vegetation types are mostly explained by the distribution of ^{137}Cs deposition (Fig. 1). However, in the spatial distribution of ^{137}Cs concentration considering the vegetation map (combined map in Fig. 4), a mosaic-like spatial variation can be observed due to the above mentioned vegetation distribution differences.

In 2016, five years after the accident, the ^{137}Cs concentration had decreased quite rapidly, with the ^{137}Cs concentrations in the litter in evergreen conifers ranging from 2.79×10^{-1} kBq kg^{-1} to 5.99×10^{2} kBq kg^{-1} (Fig. 4 and Fig. S1). At this period, ^{137}Cs concentrations in the litter of deciduous broadleaf forests were almost in the same range as those in evergreen coniferous trees (1.53×10^{-1} kBq kg^{-1} to 5.52×10^{2} kBq kg^{-1}). The rapid decrease in ^{137}Cs concentrations during the initial 5 years is attributed to the decomposition and leaching of components directly contaminated by initial deposition (including the directly polluted forest canopy) on the forest floor and the transmigration of ^{137}Cs to the soil layer. Moreover, the supply of litter fallout from the forest canopy with low ^{137}Cs concentrations resulted in a dilution effect that reduced ^{137}Cs concentrations in the litter layer. The order of decreasing rate of litter ^{137}Cs concentrations in both vegetation types were comparable with the observation in the mixed forest (Red pine and Konara Oak) and matured Japanese cedar forest in Fukushima reported by Takahashi et al. (2018). Koarashi et al. (2016) reported that evergreen conifers preserved more ^{137}Cs in the litter layer than deciduous broadleaf trees at the time of observation 2.5 years after the accident, but there was no significant difference in the ^{137}Cs concentrations in the litter layer between vegetation types. This is consistent with our estimate of concentration differences between vegetation types in 2016, although the timing differs by several years.

In 2021, 10 years after the accident, a further decrease in the litter ^{137}Cs concentration was observed in evergreen coniferous trees (range: 6.61×10^{-2} kBq kg^{-1} to 8.22×10 kBq kg^{-1}), but the rate of decrease slowed down (see also Fig. S1). In deciduous broadleaf forests, the slowdown of the decreasing trend was more obvious (1.45×10^{-1} kBq kg^{-1} to 3.60×10^{2} kBq kg^{-1}), and on average, ^{137}Cs concentrations were higher than those in evergreen conifers. This is due to the recontamination of branches and leaves of deciduous broadleaf trees via roots and higher ^{137}Cs concentration in the leaves of deciduous broadleaf trees. Kato et al. (2019) reported that ^{137}Cs concentrations in new leaves at six years after the accident showed that recontamination via roots was more significant in Konara oak than in evergreen conifers (cedar and cypress) and that the concentration was higher in Konara oak leaves after 6 years. This finding is consistent with our simulation result. This vegetation difference in the simulation strongly reflects the trend in the calibration dataset in Japanese cedar (treated as evergreen conifer forests) and Konara oak forests (treated as deciduous broadleaf forests) for FoRothCs model parameters. ^{137}Cs concentrations in Konara oak showed increasing trends (Imamura et al., 2017), while the trend was unclear for the Japanese cedar forest (Gonze et al., 2021). In the latest study by Ohashi et al. (2022), the mechanism of interspecific differences is not so simple, as some sites showed a clear increasing trend even in cedar forests, while others reached an almost steady state. However, even in the latest database, the concentration of ^{137}Cs in the leaves (trunks) of *Quercus serrata* has been consistently increasing since the accident, and the ^{137}Cs concentration in Konara oak leaves tends to be higher than that in cedar leaves in the latter half of the 2010s (Hashimoto et al., 2020c,b).

In the 10 years from 2021 to 2031, the rate of decrease in ^{137}Cs concentrations in both vegetation types is even slower than in the first 10 years after the accident, with the rate of decay generally following the physical decay of ^{137}Cs (Fig. 4 and Fig. S1). This is due to the redistribution of forest radioactive Cs in the FoRothCs model approaching dynamic equilibrium Nishina and Hayashi (2015). However, the recent model inter-comparison study showed that the simulated forest ^{137}Cs dynamics in the future period are quite different among 6 radioecological models (Hashimoto et al., 2021). So, it is important to note that there is a large uncertainty in our projection for this future period.

3.2. Controlling factors of spatio-temporal distribution of ^{137}Cs concentrations for the litter layer

In this section, to focus more on the effects of environmental factors other than deposition, we evaluated the ^{137}Cs concentration standardized by the amount of ^{137}Cs deposition (i.e., standardized ^{137}Cs concentration (m^{2} kg^{-1}) = ^{137}Cs concentration (Bq kg^{-1})/^{137}Cs deposition (Bq m^{-2})).

The simulation results show that the standardized ^{137}Cs concentration (m^{2} kg^{-1}) by deposition is correlated with the mean annual temperature (Fig. 5). A positive correlation was observed in the initial phase (0.322 and 0.305 for evergreen conifers and deciduous trees, respectively, and both were significant at $p < 0.01$); however, a negative correlation was observed for evergreen conifers in 2021 (−0.561, $p < 0.01$) (Fig. 5). The positive correlation in 2011 is attributed to the

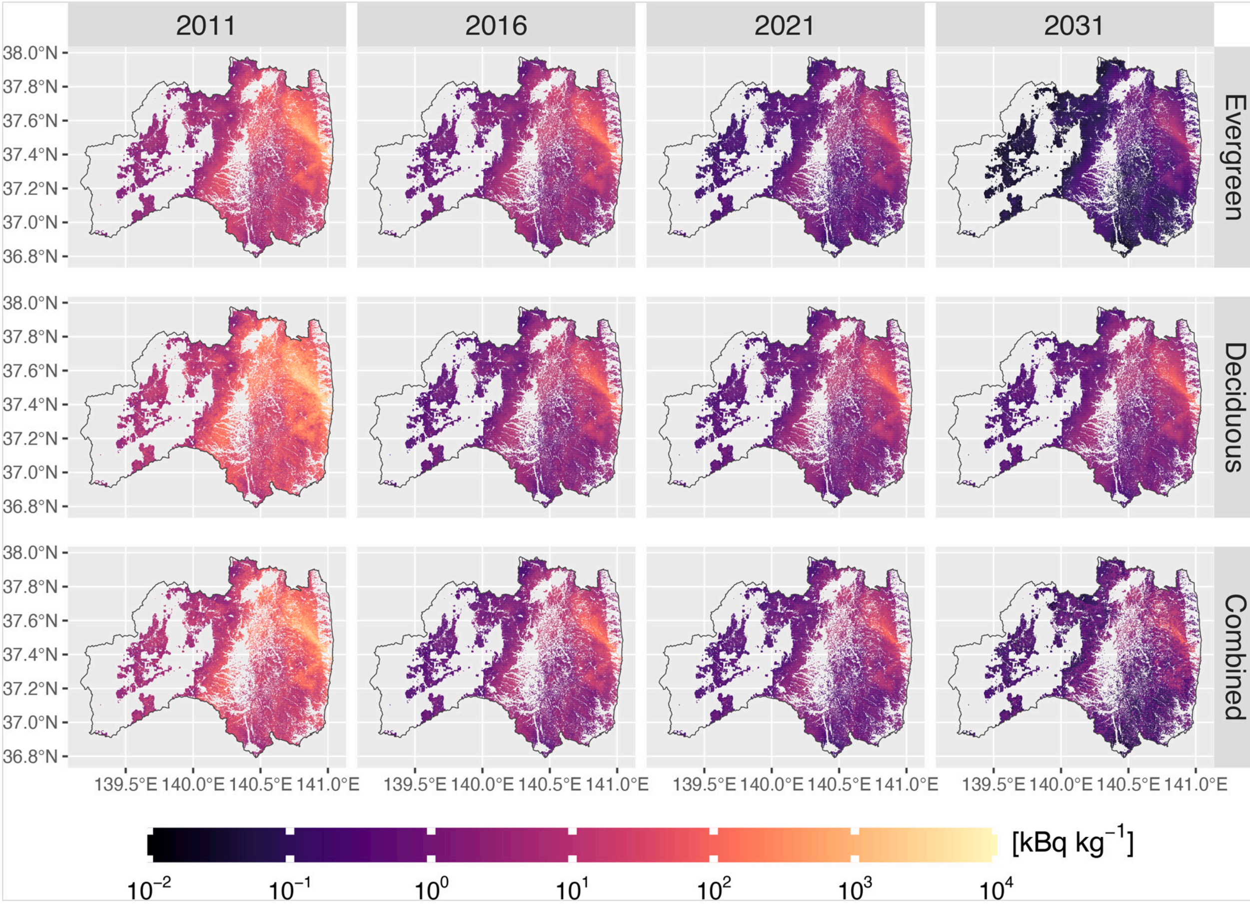

Fig. 4. Maps of simulated litter ^{137}Cs concentrations in 2011, 2016, and 2021. "Evergreen" indicates evergreen conifer forest and "Deciduous" indicates deciduous broadleaf forest. The top two rows of maps show the results of calculating the entire simulation domain as each vegetation type only. The map in "Combined" is a harmonized version of the maps in the top two rows, reflecting the vegetation distribution map (Hashimoto et al., 2020a).

difference in the amount of litter from different locations at the time of ^{137}Cs fallout. In areas with lower mean annual temperatures, litter decomposition is slower, and litter mass tends to be relatively higher; therefore, ^{137}Cs deposition per unit weight of litter is likely to be relatively smaller. This can be explained by the same mechanism in both evergreen coniferous and deciduous broadleaf forest ecosystems.

Conversely, 10 years after the accident, the correlation switched to negative for the evergreen conifer (−0.561 ($p<0.01$) in 2021, and −0.470 ($p<0.01$) in 2031). This indicates that, in evergreen coniferous forests located in the cooler region, ^{137}Cs tend to persist in the litter layer. In the model, higher ^{137}Cs concentrations are more likely to be observed in cooler sites because of lower litter decomposition activity, which allows ^{137}Cs to remain in the litter longer, and also because upward migration from the soil to the litter layer is more likely to occur as a result of longer turnover time in the litter. It has evidently been shown in laboratory experiments that under conditions of high temperature, ^{137}Cs is easily lost from in the organic layer by decomposition (Witkamp and Frank, 1970). On the other hand, litter decomposition in deciduous broadleaf forests is faster than in coniferous litter, resulting in relatively rapid decomposition of broadleaf litter even at low temperatures and, consequently, a negative correlation is not apparent as shown in Fig. 5. Although the upward movement by microorganisms into the litter in this study is always constant for each vegetation type regardless of environmental conditions, previous studies have shown that it varies with microbial biomass, vegetation, and litter site (Fukuyama and Takenaka, 2004; Huang et al., 2016; Kurihara et al., 2018a).

There was no strong correlation between annual rainfall and standardized ^{137}Cs concentrations at any time of the year (Fig. S2). The reason for this is that, in this region, it is not dry enough to inhibit decomposition activity of litter especially during the summer season. Thus, less sensitivity to annual precipitation is found in the simulation. However, this does not indicate that soil moisture is not an important factor in the ^{137}Cs dynamics of litter. In Japan's steep mountainous terrain, microtopography affects the accumulation pattern of the litter layer, with ridge tops having lower litter decomposition activity and thicker litter deposits (Koarashi et al., 2014). It should be noted that the spatial resolution of the simulations in this study does not account for changes in the soil moisture environment caused by microtopographical factors. However, microtopography does influence the accumulation pattern of litter layers in Japan's steep mountainous terrain, where litter decomposition activity is lower on ridge tops and litter deposition is thicker (e.g., Enoki et al., 1996; Yokobe et al., 2021). In terms of the spatial distribution in the watershed scale, the physical movement of litter also plays a role in the spatial redistribution of ^{137}Cs with respect to microtopography (Koarashi et al., 2014; Oda et al., 2022), and to account for this spatial heterogeneity, the model needs to address the physical movement of litter in more spatial detail and in three dimensions.

3.3. Contaminated litter as radioactive waste in Japan

In Japan, it is required that waste with ^{137}Cs concentration > 8 kBq kg^{-1} is treated as radioactive waste (formally designated waste) by the Act on Special Measures Concerning the Handling of Pollution by Radioactive Materials (Ministry of the Environment, Japan, 2011). Litter removal is one of the countermeasures for ^{137}Cs decontamination, but it is necessary to follow this law regarding the treatment of litter after the removal. Moreover, currently, many of those exceeding the

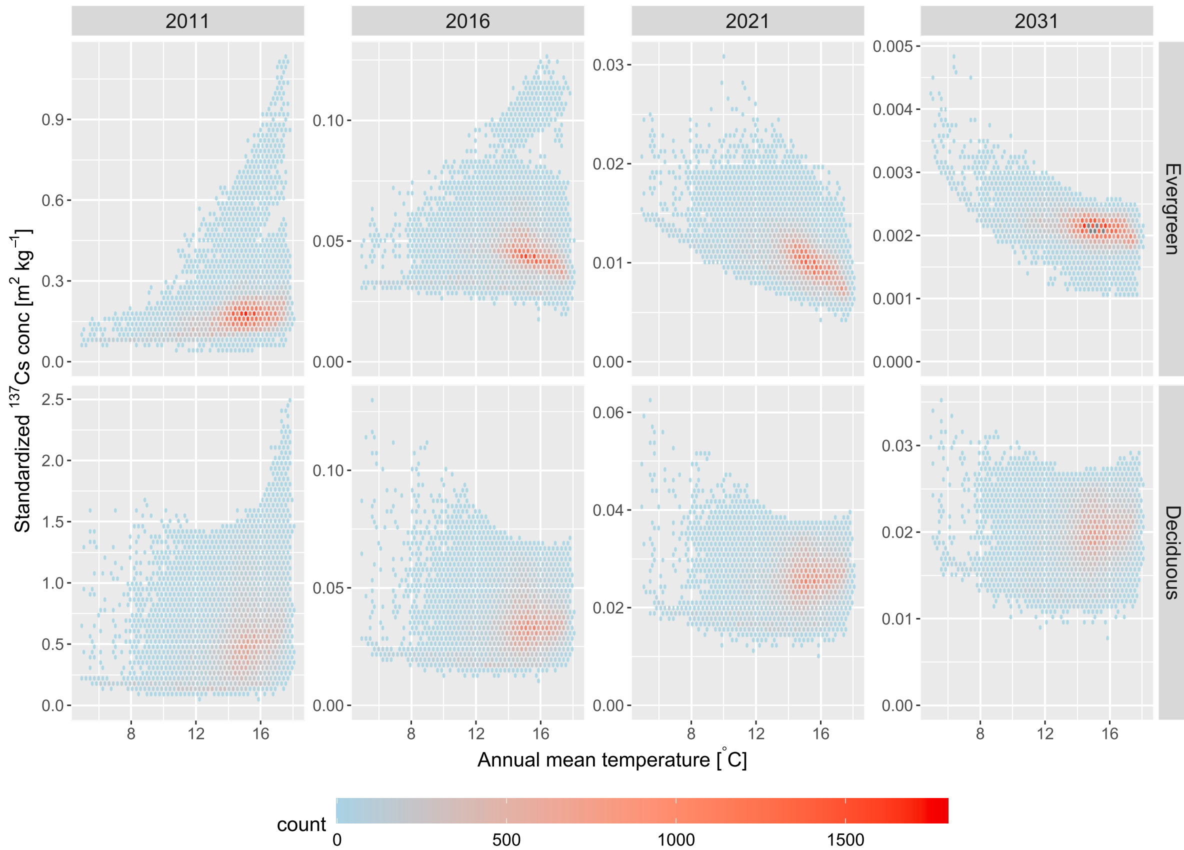

Fig. 5. Relationship between annual mean temperature and standardized ^{137}Cs concentrations in litter layer. The correlation coefficients for Evergreen are 0.322 in 2011, -0.004 in 2016, -0.561 in 2021, and -0.470 in 2031, respectively. The correlation coefficients for Deciduous are 0.305 in 2011, 0.107 in 2016, 0.198 in 2021, 0.228 in 2031, respectively. Standardized ^{137}Cs concentration [m^2 kg^{-1}] = litter layer ^{137}Cs activity concentration [Bq kg^{-1}]/^{137}Cs deposition [Bq m^{-2}].

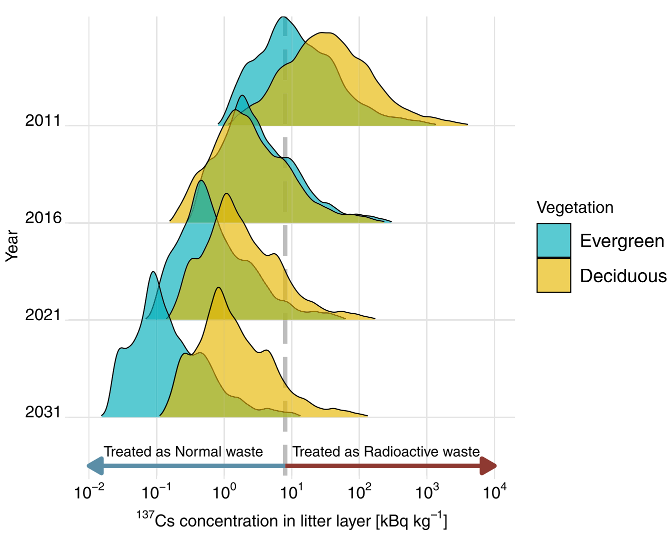

Fig. 6. Density distributions of simulated litter ^{137}Cs concentrations in each vegetation in 2011, 2016 and 2021. The gray dashed line indicates 8 kBq kg^{-1}, which is the criteria of radioactive waste in Japan.

standard values are temporarily stored in intermediate storage facilities. Since the concentration of contaminated litter relative to the standard value is important in the handling of litter, this study focuses on this standard value and analyzes the change over time in the percentage of litter exceeding the standard value (Fig. 6). As shown in Fig. 4, the litter ^{137}Cs concentrations have dramatically decreased in the two vegetation types in the first 10 years after the accident, so the percentage of area in excess of the reference value becomes smaller as time progresses. For evergreen coniferous trees, the percentage exceeding the standard value are 67.5% in 2011, 23.5% in 2016, 5.0% in 2021, and 0.7% in 2031. Conversely, because of the large influence of initial deposition in the deciduous broadleaf forest, 84.0% area in the simulated domain exceeded the standard values for radioactive waste in April 2011. Although the percentage declined steadily over the years (to the same level as evergreen conifers in 2016), approximately 13.8% of the area of deciduous broadleaf forests, more than twice the area of evergreen conifers, will be areas that exceed the standard in 2021. Simulation results show that close to 10% of the area will still exceed the standard after 20 years. This is due to the increasing contamination of hardwoods via roots, as discussed in the previous subsection.

As suggested by previous studies (Nishina and Hayashi, 2015; Cresswell et al., 2016; Hirano et al., 2016; Ayabe et al., 2017; Koarashi et al., 2020), litter removal as decontamination is effective in reducing the inventory of ^{137}Cs or the dose rate in Japanese forest ecosystems only when performed within a few years after the accident. Conversely, surface soil stripping is still an important countermeasure method, since most of the ^{137}Cs remains within 10 cm of the surface layer even today (Onda et al., 2020). At this time, it is important to treat the litter layer as nonradioactive waste from the viewpoint of waste volume reduction. However, the results of this study indicate that, in areas with high ^{137}Cs deposition, especially at higher elevations and in deciduous broadleaf forest, there is a high probability that radioactive waste-level litter layers still exist.

3.4. *Future research*

In the FoRothCs model, the litter layer is represented by a single compartment, but in reality, it contains organic matter at different stages of decomposition and distribution of ^{137}Cs changes over time (Shcheglov, 1999). The SOLVEG-II model developed by (Ota and Koarashi, 2022) distinguishes between direct litter contamination due to initial deposition and litter contamination due to redistribution via tree uptake. This might be important for improving the ^{137}Cs migration, especially in the early stages of the model. Differences in litter decomposition characteristics and pullback rates (Table S3) between the vegetation types also affect the long-term extent of ^{137}Cs contamination of the trees and thus the litter layer. These points also need to be verified in more detail with observations but are beyond the scope of this study and therefore not addressed. Further development of the litter process in the FoRothCs model is important to improve the accuracy of future predictions and our understanding of litter ^{137}Cs dynamics. In terms of validation, the annual mean temperatures of the observation points included in the database are generally biased around 8 °C to 10 °C, and there is insufficient variation for the temperature fluctuations observed in this region. Furthermore, the number of observation sites for ^{137}Cs in forest ecosystems has been decreasing with the years following the accident. To understand more accurately the dynamics of ^{137}Cs in the litter layer, regular wide-area observations over a long period of time are desirable.

Accurate estimation of biomass is essential for estimating the amount of organic matter in the litter layer since it is difficult to obtain detailed information on forest management over a wide-area. To evaluate biomass, the use of satellite monitoring should be considered in the future. Moreover, the mass of the litter layer in the FoRothCs output has not been verified at this time due to the lack of sufficient observations of litter volume over a wide-area in this region. Thus, we believe that the accuracy of the forest biomass should be validated and improved to provide more plausible estimates of the litter mass, as it is directly related to the estimation of the amount of radioactive waste.

Finally, as suggested by previous studies (Onda et al., 2020), differences in vegetation type have a significant effect on ^{137}Cs dynamics. In our simulation, the parameter of ^{137}Cs root uptake rate of deciduous broadleaf trees is 50 times higher than that of evergreen conifers (Table S3). The reason for the large differences in root uptake rates could be attributed to the different vertical fine root distributions between the tree species, whereas the surface layer of the soil is contaminated with ^{137}Cs. It had already been predicted in an early modeling study by Mahara et al. (2014) that the ^{137}Cs uptake rate via the roots in Japanese cedar was slower than that in Konara oak, mainly because the Japanese cedar tree distributes few fine roots within 10 cm of depth. Conversely, Japanese oak is known to distribute many highly active fine roots within 10 cm of the surface (Makita et al., 2011) and thus is more likely to uptake ^{137}Cs due to the contamination being concentrated at the surface layer (usually up to 5 cm). Such differences in the depth distribution of nutrient acquisition between evergreen conifers and deciduous broadleaf trees have also been observed in the acquisition of nitrate ions (Morikawa et al., 2022). Even nearly 10 years after the accident, ^{137}Cs has remained within the 10-cm layer in many forests (Manaka et al., 2022). However, in the next few decades, it will gradually move further down the layers due to biological disturbance. To account for this process, the simulated litter ^{137}Cs concentrations in 2031 could be higher than the current result for evergreen conifer forests, and conversely lower for deciduous broadleaf forests. Since the current FoRothCs treats soils in bulk, we consider that reproducing the ^{137}Cs and root depth distribution in the model will be an important factor for the forecast in the next several decades.

4. Conclusion remarks

Using the FoRothCs model, we have successfully simulated ^{137}Cs concentrations in the litter layer contaminated by the FNDPP accident with acceptable accuracy for the last 10 years of observations. The evaluation of litter layer ^{137}Cs concentration is possible because FoRothCs couples biomass growth and organic decomposition with ^{137}Cs dynamics. Our simulation suggested that the area with lower annual air temperature (i.e., high-altitude area) tends to retain ^{137}Cs in the litter layer, since the decomposition activity of the litter layer is low in such areas. We found that environmental factors such as vegetation type have a strong influence on spatiotemporal variations in ^{137}Cs concentration in the litter layer. Our results suggest that differences in ^{137}Cs litter contamination due to the differences in vegetation type will become important even in the later stages of contamination after 10 years. Continued attention should be paid to the distribution of vegetation within the watershed when considering future ^{137}Cs budgets in the watershed.

Despite the model limitation and uncertainty of detailed forest stand information in each grid, the prediction of regional litter ^{137}Cs concentrations by FoRothCs showed good agreements with the observations (rho = 0.690 and 0.663 in each vegetation type) and was useful in determining the degree of contamination of the litter layer. Such information is useful for not only for predicting litter concentrations in forest ecosystems but also for predicting the concentration of radioactive materials in plants and animals in forests and watersheds. For example, it has been reported that ^{137}Cs concentrations in the litter layer are good indicators of contamination for some species of saprophytic fungi (Komatsu et al., 2019, 2021) and that the ^{137}Cs concentration in the leaves of Koshiabura (*Eleutherococcus sciadophylloides*), which is used as an edible wild plant, is strongly correlated with the ^{137}Cs concentrations in the litter layer (Kiyono et al., 2019). For the river ecosystem, the estimation of ^{137}Cs concentrations in the litter layer over a wide-area, coupled with models such as river transport models (Kurikami et al., 2019; Sakuma et al., 2022), will contribute to a better understanding of the future behavior of ^{137}Cs in the entire watershed.

CRediT authorship contribution statement

Kazuya Nishina: Conceptualization of this study, Methodology, Software. **Seiji Hayashi:** Software, review draft. **Shoji Hashimoto:** Methodology, Data curation, review draft. **Toshiya Matsuura:** Data curation, review draft.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Kazuya Nishina reports financial support was provided by JSPS KAKENHI.

Data availability

The data that has been used is confidential.

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Appendix A. Supplementary material

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.envpol.2023.121605.

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