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Radiocesium leaching from litter during rainstorms in the Fukushima broadleaf forest

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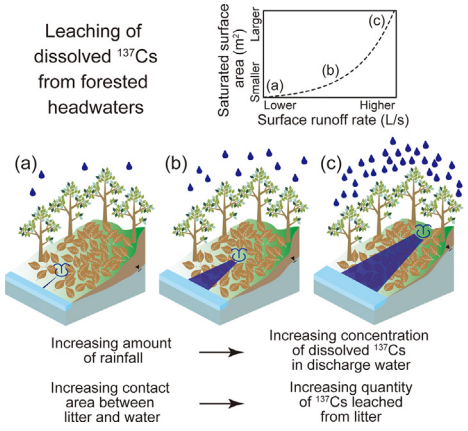
HIGHLIGHTS

- Leaching of ^{137}Cs released from the Fukushima accident was examined.
- Conducted leaching tests for ^{137}Cs in broadleaf litter and saturated runoff.
- The area of contact between runoff water and litter increased during rainstorms.
- Dissolved ^{137}Cs in runoff water could be explained by leaching from litter.
- The presence of contaminated litter in headwaters is related to elevated ^{137}Cs.

GRAPHICAL ABSTRACT



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ABSTRACT

Forests are important sources of dissolved radiocesium (^{137}Cs) discharge downstream. To improve understanding of dissolved ^{137}Cs discharge processes during rainstorms, we investigated the relationship between rainfall-runoff hydrological processes and the discharge of ^{137}Cs leached from litter. Leaching tests were conducted with broadleaf litter collected in the area where saturated overland flow was generated during rainstorms in a broadleaf-tree-dominated forest. According to the leaching test results, the ^{137}Cs leaching rate was higher in the early stage of the test and decreased afterward. There was no significant difference in the overall results between the agitation and non-agitation cases. The ^{137}Cs leaching rate from litter after the 24-h test was up to 33.7%. A large proportion of the original ^{137}Cs activity was present even after the tests, as leaching from litter during rainstorms in the headwater area could be an additional source of dissolved ^{137}Cs in the stream water. If mixing of ^{137}Cs originating from groundwater, soil water, and rainfall with the hydrological processes is assumed, differences between the observed and estimated ^{137}Cs in the surface runoff water became larger under high flow conditions. This analysis indicates additional ^{137}Cs loading on surface runoff water during rainstorms, where saturated surface area can expand as the surface runoff rate increases. Contact area between surface runoff and litter accumulated on the forest floor should increase and accelerate ^{137}Cs leaching from the litter. Therefore, ^{137}Cs leaching in the saturated surface area that is temporarily formed during rainstorms can play a principal

Abbreviations: FDNPP, Fukushima Dai-ichi Nuclear Power Plant; IPL, Isotope Products Laboratories.

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role in dissolved ^{137}Cs discharge during rainfall-runoff events. Contaminated litter in the temporally saturated region of forested headwaters is an important factor contributing to elevated levels of dissolved ^{137}Cs during rainstorms in the Fukushima area.

1. Introduction

The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, triggered by a huge earthquake and tsunami on March 11, 2011, released massive amounts of radionuclides into the environment, including radiocesium (^{137}Cs), (Chino et al., 2011; Hirose, 2012), which were redistributed in the terrestrial environment (Onda et al., 2020). The terrestrial area affected by the FDNPP accident is predominantly covered by forests (Hashimoto et al., 2012; Kato et al., 2018). A large fraction of ^{137}Cs discharged from the FDNPP into the forests is intercepted by the forest canopy (leaves and branches) before deposition onto the ground surface (Kato et al., 2012). Intercepted ^{137}Cs is transferred from forest canopy to floor through litterfall, throughfall, and stemflow (Teramage et al., 2014; Calmon et al., 2015); hence, forest canopy delays the transfer of ^{137}Cs from the atmosphere to the forest soil (Kato et al., 2012). Such transferred ^{137}Cs is affected by two main processes: 1) absorption and fixation among the clay minerals of the surface soil layer (Cremers et al., 1988; Takahashi et al., 2015); 2) discharge downstream through soil erosion and water runoff (Chartin et al., 2013; Iwagami et al., 2017a).

Principal factor of ^{137}Cs downstream transport is the discharge of suspended sediment (particulate matter) carried by rivers (e.g., Taniguchi et al., 2019). For example, Yamashiki et al. (2014) investigated the flux of ^{137}Cs in the Abukuma Basin from August 2011 to May 2012, which is the largest river system in the FDNPP-accident-affected region. They concluded that 84%–92% of total ^{137}Cs discharge was in particulate form. Dissolved ^{137}Cs has also been inferred as another important phase during radionuclide downstream discharge (Iwagami et al., 2017a). Yoshimura et al. (2015) studied both the particulate and dissolved phases of ^{137}Cs in river water at 30 sites close to the FDNPP during December 2012 and found a significant correlation between the ^{137}Cs concentration and catchment inventory.

Several studies have monitored ^{137}Cs discharge (e.g., Iwagami et al., 2019a; Nakanishi and Sakuma, 2019; Taniguchi et al., 2019). They commonly concluded that ^{137}Cs concentration in the river or stream water rapidly declined by one or two orders of magnitude within approximately one year after the FDNPP accident, before slowing down.

Temporally increasing trends in both particulate and dissolved ^{137}Cs concentration in surface runoff water during rainstorms have been observed by previous studies focusing on heavier-contaminated forests (^{137}Cs deposition <3.0 MBq/m^2, catchment area <30 km^2) (e.g., Ueda et al., 2013; Tsuji et al., 2016; Iwagami et al., 2017b), although some studies focusing on the less-contaminated (<0.1 MB/m^2) larger catchments (>500 km^2) (e.g., Nagao et al., 2013) observed a decrease in dissolved ^{137}Cs concentration during rainstorms. This is similar to the characteristics of the region affected by the Chernobyl accident (Nylen and Grip, 1997). Increase in ^{137}Cs discharge in the particulate phase during rainstorms has been explained as a result of an increase in suspended sediment concentration with a high level of ^{137}Cs due to soil erosion, especially in the surface layer of the ground (Nagao et al., 2013). However, causes of increased dissolved ^{137}Cs discharge concentrations during rainstorms continue to be studied.

Within two years of the FDNPP accident, dissolved ^{137}Cs concentration in the surface runoff water was reported to be more than one order of magnitude higher during rainstorms than when there was no rainfall in contaminated headwater catchments (Iwagami et al., 2017b). However, 2–4 years post-accident, increased concentration of dissolved ^{137}Cs in the river (catchment area: 21 km^2) during rainstorms was approximately twice as large as that in the absence of rainfall (Tsuji et al., 2016). In the case of contaminated headwater catchment (catchment area: less than 0.05 km^2), there was no clear change in the dissolved ^{137}Cs concentration in the surface runoff water to increase by one order of magnitude during rainstorms even 4–5 years after the incident (Iwagami et al., 2019b).

Moreover, Sakuma et al. (2019) applied a distribution coefficient (K_d) absorption/desorption model to simulate the concentration of dissolved ^{137}Cs in both baseflow and high-flow conditions targeting a forested catchment from January 2014 to December 2015 based on the knowledge that dissolved ^{137}Cs concentrations in rivers were correlated with both catchment ^{137}Cs inventory (e.g., Tsuji et al., 2014) and ^{137}Cs concentrations in suspended solids (e.g., Yoshimura et al., 2015). As a result of calibration with the data reported by Tsuji et al. (2016), it was found that the mean dissolved ^{137}Cs concentration in rivers during baseflow could be reproduced, whereas seasonal variability (Tsuji et al., 2016) and peak concentrations of the dissolved ^{137}Cs in rivers during rainstorms (e.g., Iwagami et al., 2019b) could not be reproduced well. They concluded that the results that could not be reproduced were likely related to factors such as leaching of ^{137}Cs from organic matter in forest litter layers; these were unaccounted for in the model. This could be an indication that the equilibrium between particulate and dissolved ^{137}Cs was not achieved during rainstorms. Based on the interpretation of Sakuma et al. (2019), it is assumed that the discrepancy between observed and simulated dissolved ^{137}Cs concentrations in river waters in more contaminated and forested headwaters, where the litter layer is more developed, will be larger in the current model. Therefore, investigating mechanisms behind increased dissolved ^{137}Cs concentration in surface runoff water during rainstorms in contaminated and forested headwaters is important, and will positively influence environmental safety while assisting with improving the model.

Sakakibara et al. (2019) investigated the rainfall-runoff processes in a forested headwater area in Fukushima and concluded that groundwater discharge was the main component, with more than 50% contribution to the surface runoff water during rainstorms. Considering rainfall-runoff processes, dissolved ^{137}Cs in groundwater is a possible contributor to discharge during rainstorms. However, Iwagami et al. (2017b) and Iwagami et al. (2019b) observed more than one order of magnitude lower ^{137}Cs concentrations in groundwater than those in surface runoff water in contaminated headwater catchments in August 2011 and from July 2015 to November 2016, respectively. Considering lower contribution of rainfall to surface runoff water during a rainstorm (Sakakibara et al., 2019) and firmly absorbed ^{137}Cs on the soil particles (Cremers et al., 1988; Fukushi et al., 2014), another source of ^{137}Cs could be attributed to discharge by leaching from the litter on the forest floor (e.g., Tsuji et al., 2016; Iwagami et al., 2019b). This is because the litter layer is one of the most contaminated components of the environment surrounding the FDNPP area (Teramage et al., 2014).

^{137}Cs concentrations in both litters in streams and forests have been reported (e.g., Murakami et al., 2014; Sakai et al., 2016a; Sakai et al., 2016b). Murakami et al. (2014) reported one to two orders of magnitude higher ^{137}Cs content in forest litter, and Sakai et al. (2016a) observed four times higher ^{137}Cs content in forest litter than in stream litter in the Fukushima area. This is suggested to be the result of ^{137}Cs leaching from the litter by water. Sakai et al. (2015) conducted a ^{137}Cs leaching test using fresh conifer litter affected by the FDNPP accident that spanned several days and revealed leaching of ^{137}Cs from litter

and corresponding leaching rates. Kurihara et al. (2020) and Sakuma et al. (2021) implemented ^{137}Cs leaching tests using both conifer and broadleaf litter. They indicated that ^{137}Cs was more likely leached from broadleaf litter than from conifer litters, but ^{137}Cs in well-decomposed litter was less leachable in water.

Considering the findings of the above-described previous studies, two points should be considered for further interpretation. First, the fact that dissolved ^{137}Cs in the surface runoff water during rainstorms differs depending on the location and time elapsed after the FDNPP accident warrants further investigation. Additionally, concentration of dissolved ^{137}Cs in the surface runoff water during rainstorms has not been well reproduced using the current model because of unconsidered factors such as ^{137}Cs leaching from organic matter (Sakuma et al., 2019). Specifically, it is not well understood why such a difference in time and location occurs, and especially why the elevated dissolved ^{137}Cs is released during rainstorms even after a long period since the accident in the forested headwater catchment. Second, in terms of the elevated levels of dissolved ^{137}Cs in the surface runoff water during a rainstorm, there is a unified view that this is caused by ^{137}Cs leaching from litter; however, few studies have considered the loading process of ^{137}Cs leached from litter into the surface runoff water with respect to the hydrological processes in the forested headwaters. Consequently, the objective of this study is to improve the understanding of dissolved ^{137}Cs discharge from forested headwaters. Therefore, this study investigates the factors that connect the leached ^{137}Cs from forest litter and elevated dissolved ^{137}Cs concentrations in the surface runoff water during rainstorms. This study examines the ^{137}Cs leaching characteristics (including leaching rate) from forest litter and discusses the processes that lead to an increase in dissolved ^{137}Cs in the surface runoff water during rainstorms in terms of the ^{137}Cs leaching from litter and the hydrological processes, focusing on the contaminated and forested headwater catchment.

2. Study site

The study area is a small headwater catchment (Fig. 1; area: 0.048 km^2; elevation: 555–672 m; geology: weathered and fractured granite) in Yamakiya District, Kawamata City, Fukushima Prefecture, Japan, the same catchment investigated by Sakakibara et al. (2019) and Iwagami et al. (2019b). The entire land surface is covered by forest (29% conifer, 71% broadleaf). The broadleaf forest consists of deciduous broadleaf trees, which are mainly Japanese Konara oak; therefore, many deciduous leaves are found on the ground surface all-year. The study area is in a temperate humid climate region with an annual average air temperature and precipitation of 10.3 °C and 1350 mm, respectively. Heavy rainfall events greater than 100 mm precipitation/event occasionally occur in the rainy (June–July) and typhoon (August–September) seasons.

The study area is approximately 35 km northwest of the FDNPP and has a ^{137}Cs deposition density of 1.2 MBq/m^2 (Kato et al., 2019). This catchment represents one of the headwaters of Abukuma River, the sixth-longest river in Japan, and flows through large cities such as Fukushima and Koriyama. Water from the catchment is linked to resources for drinking, daily use, agriculture, and industry in the Abukuma River watershed, necessitating a study of radionuclide migration processes in this catchment. A more detailed explanation of the study area can be found in the studies by Sakakibara et al. (2019) and Iwagami et al. (2019b).

3. Materials and methods

3.1. Litter sampling and ^{137}Cs analysis of litter

Litter sampling was conducted on November 17, 2015, in the catchment area (Figs. 1 and 2-a). Sampling location was selected to be where the saturated overland flow was generated only during a rainstorm (Fig. 2-b: photo in the rainless period, Fig. 2-c: photo during a rainstorm). November is in the autumn season, and hence many leaves of broadleaf trees fall onto the forest floor in a single day. According to rain data observed in the catchment (Sakakibara et al., 2019), total rainfall was 0 and 0.4 mm/day on November 16 and 17 (until litter sampling), respectively. Thus, dry and undecomposed litter accumulated on the uppermost layer of the ground surface. Approximately 1 kg of broadleaf litter was sampled (Fig. 2-d). Decomposed litter was found in deeper layers of litter. However, decomposed litter was not collected in this study because the dissolved ^{137}Cs concentration leached from the decomposed litter layer could be included in the results of pore water in the mineral layer of Iwagami et al. (2019b) (see Section 3.3). Pore water is defined as “soil water” in the following sections of the manuscript. This study assumed that the cause of the increase in the dissolved ^{137}Cs concentration in the surface runoff water during rainstorms is leaching of ^{137}Cs from the litter on the saturated surface area that temporarily forms during rainstorms.

The sampled litter was gently, but rapidly wiped using clean paper with ultra-pure water to remove attached soil. Because of the wetness variability in litter samples, they were dried in an incubator at 60 °C for seven days. To determine the levels of ^{137}Cs in the sampled litter,

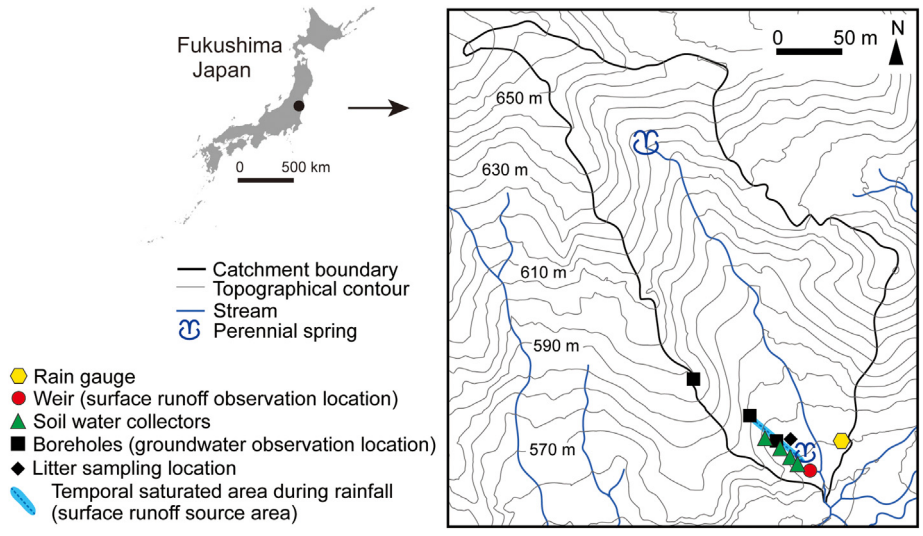

Fig. 1. Study area map with topographical information and locations of the temporal saturated surface area and installed equipment by Sakakibara et al. (2019) and Iwagami et al. (2019b).

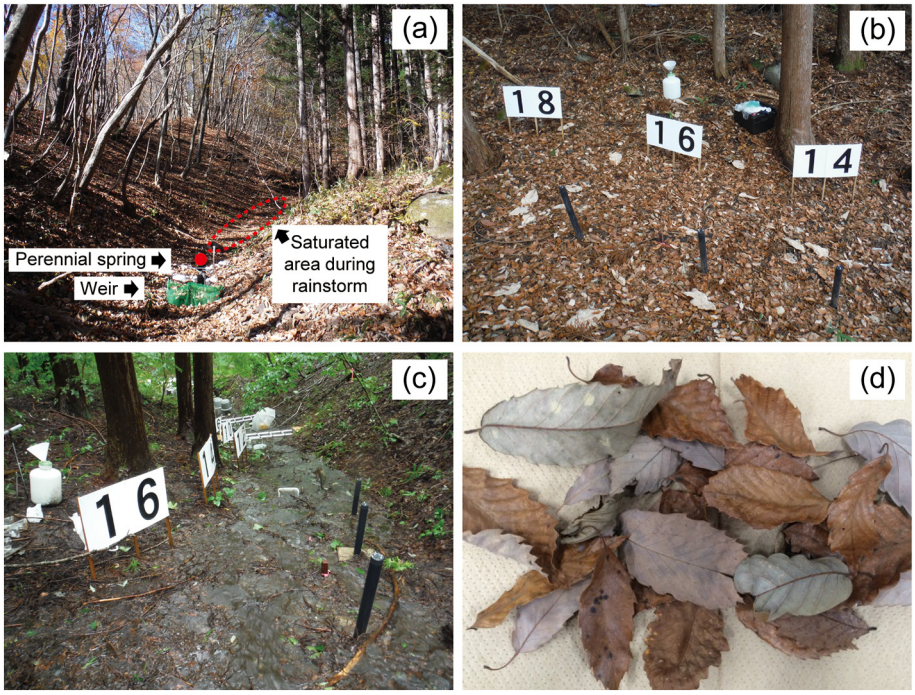

Fig. 2. Photos of (a) the study catchment, (b) a condition of litter sampling location before rainstorms, (c) a condition of litter sampling location during a rainstorm, and (d) the sampled litter (Japanese Konara oak). During a rainstorm, a saturation area of up to 40 m long and 2 m wide is formed in the upstream direction, and saturated overland flow generates as shown in photo (c).

samples were crushed using a mixer. The ^{137}Cs concentrations were analyzed using gamma-ray spectrometry with germanium semiconductor detectors (GC4019, Canberra, USA) at the University of Tsukuba, Japan. The spectrometers were calibrated with a gamma-ray-certified reference material (source number: 752-69) containing ^{137}Cs from Isotope Products Laboratories (IPL). Physical decay of ^{137}Cs was corrected based on the sampling date. The ^{137}Cs analysis of the litter was repeated 10 times to obtain the average value and standard deviation of the measurements.

3.2. Leaching test and ^{137}Cs analysis of leaching solution

Leaching tests were conducted to examine the ^{137}Cs leaching rate and its temporal characteristics from the litter affected by the FDNPP accident. Detailed procedure and conditions of the tests are summarized in Fig. 3 and Table 1, respectively.

Crushing treatment for litter samples was not performed to reproduce the field conditions. Sakai et al. (2015) used stream water at the headwater catchment affected by the FDNPP accident and distilled water to confirm the differences in the water composition in the analyses of the ^{137}Cs leaching quantities from litter samples. They reported minor differences between the stream and distilled water. Hence, distilled water was used for the leaching tests in this study. Litter samples (4 g) and distilled water (500 mL) were placed inside clean 1-L glass beakers. Leaching tests were performed in two ways in the laboratory at a constant room temperature (22 °C, assuming field condition in summer), one with non-agitation (0 rpm), and the other with agitation at 100 rpm using a mechanically controlled agitator (RW20 Digital, AS ONE, Japan). The leaching test was conducted in two ways because we assumed the process of surface runoff generation in forested headwaters. The non-agitation case assumed the process of saturated surface area generation by the rise of groundwater table level above the ground surface, and the agitation case assumed the process of overland flow generation from the saturated surface area. To examine ^{137}Cs leaching characteristics in the order of hours, as in a rainfall event, non-agitation (0 rpm) and agitation (100 rpm) periods were set as 0.5, 1, 2, 6, 12, and 24 h. Each test was repeated five times to obtain the average value and standard deviation of the measurement results.

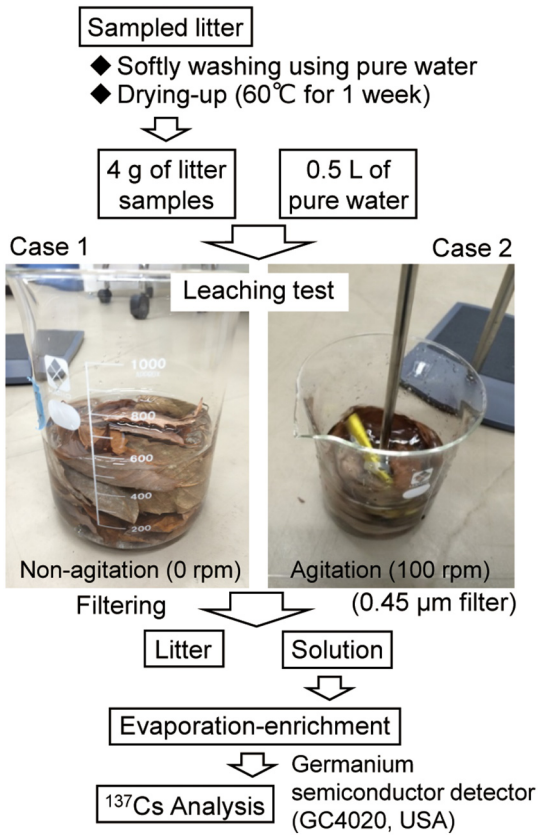

Fig. 3. ^{137}Cs leaching tests processes using sampled litter.

Table 1
Conditions of ^{137}Cs leaching tests using the sampled litter.

	Case1	Case2
Agitation speed (rpm[a])	0	100
Litter sample	Dried broadleaf litters	
Litter[b]-water[c] ratio	4 g/0.5 L	
Test period (hours)	0.5, 1, 2, 6, 12, 24	
Number of replicates	5 for each	

[a] rpm: rotations per minute.
[b] Japanese Konara oak.
[c] Pure water (EC < 0.5 μS/cm).

After each test, the litter samples were removed from the beaker via filtering using a 0.45-μm membrane filter (ADVANTEC, Japan). Electrical conductivity (EC) of the leaching solution was measured using a compact EC meter (EC-22, Horiba, Japan) to confirm the release of specific elements. The leaching solution was then evaporated to approximately 200 mL using an induction heater to enrich the dissolved ^{137}Cs concentration. The ^{137}Cs in the solution was determined in the same way using the same analyzer as that used for the litter samples. Moreover, the ^{137}Cs leaching rate (μBq/g/s) from the sampled litter was calculated as ^{137}Cs leached from a unit of litter per unit of time. The ^{137}Cs leaching rate (percentage of ^{137}Cs leached) was calculated as follows: ^{137}Cs in the solution (leached from litter) divided by ^{137}Cs in the litter used in the test.

3.3. Application to rainfall-runoff phenomena

To discuss why the dissolved ^{137}Cs concentration in the surface runoff water increases during rainstorms, the results of leaching tests were applied to some rainfall-runoff phenomena. Hydrological data (i.e., surface runoff rate; precipitation) and dissolved ^{137}Cs data in water were not observed in this study; therefore, we referred to the results of the following two studies: Sakakibara et al. (2019) and Iwagami et al. (2019b). The study sites of Sakakibara et al. (2019) and Iwagami et al. (2019b) had the same catchment as in this study (Fig. 1). They observed surface runoff water just downstream of the perennial spring. Soil water (subsurface water adjacent to the ground surface, including water in decomposed litter layers), groundwater (water in the saturated subsurface area), and rainwater were also observed in the catchment. Sakakibara et al. (2019) conducted hydrological observations and hydrograph separation focusing on three rainstorm events (July 15–17, 2015; August 22–24, 2016; and August 29–31, 2016) using non-radioactive and conservative tracers (Cl^- and SF_6). Sakakibara et al. (2019) inferred that surface runoff water during rainstorms can be explained as a mixture of rainwater (gross rainfall), soil water, and groundwater components that each component is called "endmember of surface runoff water" in this study. They also showed the contribution of each endmember to surface runoff water. Iwagami et al. (2019b) measured ^{137}Cs concentrations in environmental waters, including surface runoff water, soil water, and groundwater, in the same catchment as this study. They also observed the ^{137}Cs concentration in surface runoff water during rainstorms, focusing on the same stream and the same rainstorm events as Sakakibara et al. (2019). The ^{137}Cs concentration of gross rainfall at approximately 17 km from the study area in 2015 was reported to be below the detection limit (<0.37 Bq/L) by Tsuji et al. (2016). The detection limit depending on sample amount was slightly high; however, considering the fact that the ^{137}Cs concentration of gross rainfall decreased rapidly in the first several months after the FDNPP accident (Kato et al., 2012), this study assumes that the gross rainfall does not contain ^{137}Cs. Using the reported and assumed results, except for the ^{137}Cs data in surface runoff water, discharge rate (Bq/s) of dissolved ^{137}Cs concentration through surface runoff water was calculated. The differences between the observed (Iwagami et al., 2019b) and calculated (this study) values were determined.

Calculated ^{137}Cs concentration in the surface runoff water was taken as the integrated ^{137}Cs concentration in each component (rainwater, soil water, and groundwater; Table 2) without the leached ^{137}Cs from the litter accumulated on the ground surface. Therefore, if the observed value was larger than the calculated value, the difference could be considered as ^{137}Cs leaching from the litter. The litter amount was estimated by compensating for the difference between the observed and calculated values. First, the elapsed time from the start of the rainfall to the water sampling was determined because the leaching rate depended on the elapsed time subsequent to the wetting of the litter (see Section 4.1). Second, average ^{137}Cs leaching rate per unit of litter (Bq/g/s) was calculated based on the leaching test results and elapsed time from the start of the rainfall to water sampling. Finally, the amount of litter was determined. It was assumed that the litter deposited on the ground before the rainstorm was dry. If the litter was wet, the amount of litter estimated in this study would have been underestimated compared to the actual value.

Table 2
^{137}Cs concentration in rainwater, soil water, and groundwater in the studied catchment area reported by previous studies.

Water	n	^{137}Cs (Bq/L)	Reference
Rainwater	–	Assumed not contained	Kato et al. (2012), Tsuji et al. (2016)
Soil water	38	0.05 ± 0.03	Iwagami et al. (2019b)
Groundwater	19	0.0009 ± 0.0004	Iwagami et al. (2019b)

The error is a standard deviation.

There are two other points to be noted. First, water samples collected by Sakakibara et al. (2019) and Iwagami et al. (2019b) were used as the same water samples in this study only if differences in their collection times were within 60 min. These two studies were conducted simultaneously, focusing on the same rainstorm events, but they did not collect water samples simultaneously. Second, the ^{137}Cs concentration in throughfall contained a higher concentration of ^{137}Cs (e.g., Teramage et al., 2014); however, ^{137}Cs in throughfall was not considered in this study. We made this decision based on the hydrological processes in the study area, due to which the rainwater component does not largely contribute to the surface runoff water because most rainwater infiltrates into the soil layer (Sakakibara et al., 2019). To verify this, ratio of the saturated surface area to the total catchment area was considered. The maximum saturated surface area was estimated to be 80 m^2 (40 m long and 2 m wide), whereas the total catchment area was 48,000 m^2. The ratio was <0.002; therefore, the contribution of ^{137}Cs in throughfall to the surface runoff water was negligible.

3.4. Quantification of expansion of saturated surface area during rainstorms

To estimate the area (m^2) of the saturated ground surface during rainstorms, an interval camera was installed in the study area, and surface condition was recorded with a 10-min time resolution. Every interval photo included signboards showing the distance (m) from the fixed surface runoff observation point; therefore, where the saturated surface flow started was recorded. Additionally, during several rainfall events, the length and width of the saturated surface flow were directly measured in situ to validate the saturated surface area through interval photos. The area of the saturated surface area was estimated using geometric calculation tools in ArcGIS with the 1-meter space resolution digital elevation model. The estimation would include a 20%–30% error because of the lack of observed saturation data of the ground surface. In total, 11 cases were examined, and the surface runoff rate in each case was obtained from the results of Sakakibara et al. (2019).

4. Results and discussion

4.1. Leaching characteristics of ^{137}Cs from litter

The average value of analyzed ^{137}Cs concentration in the litter from 10 replicates was 1430 Bq/kg, with a standard deviation of 260 Bq/kg. The average ^{137}Cs concentration (1430 Bq/kg) was used as the ^{137}Cs concentration in the sampled litter in the following sections.

The relationships between the leaching test period, EC (a), and levels of ^{137}Cs (b) in the leaching solution (0.5 L) are presented in Fig. 4. EC of the leaching solution was within the range from 7.0 to 53.2 μS/cm and from 8.0 to 50.0 μS/cm in the cases of non-agitation (0 rpm) and 100-rpm agitation, respectively. Additionally, the EC of the leaching solution increased as the test period increased in both cases. This might indicate that some solutes were loaded in distilled water from the litter samples. The ^{137}Cs activity of the leaching solution also became larger as the test period lengthened, with values ranging from 0.21 (0.5-h test) to 1.93 Bq (24-h test) and from 0.35 (0.5-h test) to 1.54 Bq (24-h test) in the cases of non-agitation and 100-rpm agitation, respectively.

Leaching rate (the percentage of ^{137}Cs leached: %) of ^{137}Cs from the litter samples ranged from 3.6% to 33.7% and from 6.0% to 30.5% in cases of non-agitation (0 rpm) and 100-rpm agitation, respectively. Even 24 h after the leaching test started, the leaching rate of ^{137}Cs from the litter samples was as high as 33.7%. Sakai et al. (2015) found that the leaching rate of ^{137}Cs was approximately 30% of the original ^{137}Cs activity of litter by implementing a 1-day leaching test at 21 °C using fresh cedar litter. Conversely, Kurihara et al. (2020) reported less than 5% of ^{137}Cs leaching rate after a 1-day leaching test at 4 °C using broadleaf litter. Our results (leaching rate: approximately 30%) were similar to those of Sakai et al. (2015), whereas there was a large gap between our results and those of Kurihara et al. (2020). This may be due to the difference in the temperature of the leaching test and the resulting difference in the degradation process of liters during the experiment, as Nakanishi and Sakuma (2019) reported the temperature dependency of dissolved ^{137}Cs concentration in river water. Our study focused on ^{137}Cs leaching from dry litter on the ground surface during summer rainstorms. Therefore, the results of our leaching test using dry litter and under 22 °C conditions could be important based on the assumptions of this study. However, a recent study (Sakuma et al., 2021) using wet litter after a litterbag experiment reported that the leaching rate was less than 10% even under 20 °C leaching tests. Therefore, more studies under a variety of conditions are a subject of future work for a unified view of the ^{137}Cs leaching characteristics from litter.

The relationship between leaching test period and leaching rate of ^{137}Cs from the litter (μBq/g/s) is presented in Fig. 5 with a regression

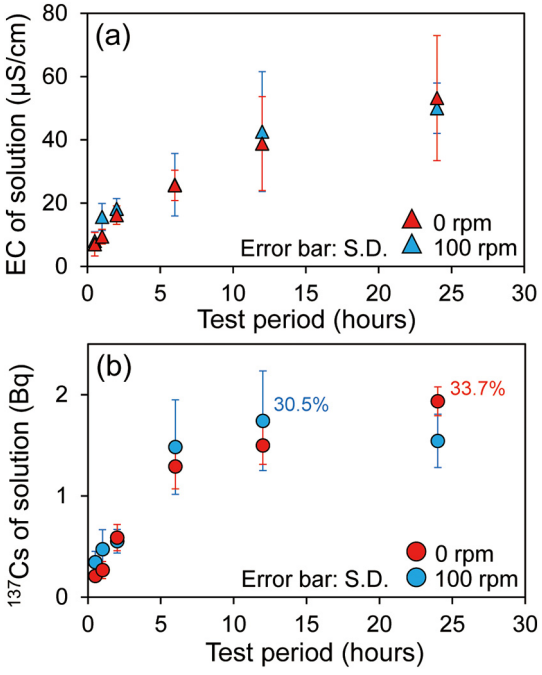

Fig. 4. Relationships between leaching test period and (a) EC, and (b) ^{137}Cs level of the leaching solution (0.5 L). Values in panel b are leaching rates (percentage of ^{137}Cs leached from the litter sample).

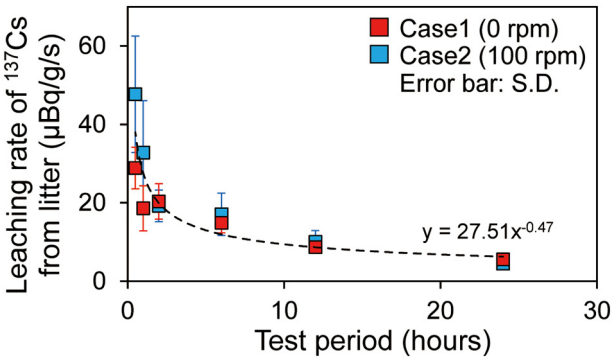

Fig. 5. Relationship between the leaching test period and leaching rate of ^{137}Cs from a unit of litter.

curve (a power approximation) using both the 0-rpm and 100-rpm data. This suggests that ^{137}Cs rapidly leached from the litter when the litter became wet. This interpretation was supported by Kurihara et al. (2020) and Sakuma et al. (2021), who inferred immediate ^{137}Cs leaching after the start of soaking in water. Therefore, when ^{137}Cs leaching from litter during a rainstorm is considered, the elapsed time from the start of the rainfall is crucial.

4.2. Estimated contribution of litter on dissolved ^{137}Cs discharge during rainstorms

Table 3 includes the surface runoff rate, endmember component contributions (rainwater, soil water, and groundwater) to surface runoff water, and the ^{137}Cs concentration in the surface runoff water during the three rainfall events reported in previous studies. Original results of this study, which are the calculated ^{137}Cs concentration in the surface runoff water and estimated litter amount that compensates for the difference between the observed and calculated ^{137}Cs discharge, are shown in Table 3.

Observed ^{137}Cs discharge rate (Bq/s) was between 0.001 and 0.093 Bq/s. The minimum and maximum ^{137}Cs discharge rates were observed at 13:00 on August 22, 2016, and at 12:00 on August 30, 2016, respectively. The former timing was just before rainfall with a surface runoff rate of 0.14 L/s, and the latter was during heavy rainfall with a surface runoff rate of 2.28 L/s. This indicates that the ^{137}Cs discharge rate increased during rainfall events. Data from 13:00 on August 22, 2016, were eliminated from further discussion because it was inappropriate to discuss the contribution of ^{137}Cs leached to the ^{137}Cs discharge rate during a rainstorm. All other data were collected during or after rainfall, with the elapsed time from the start of the rainfall between 1 and 64 h. The calculated ^{137}Cs discharge rates (Bq/s) lie between 0.004 and 0.028 Bq/s. Additionally, the difference between the observed and calculated ^{137}Cs discharge rates was 0.002–0.066 Bq/s. Finally, to compensate for this difference, litter mass was estimated to be between 320 and 13,180 g based on the ^{137}Cs leaching rate (Fig. 5), considering the elapsed time from the start of the rainfall.

Uncertainty exists in the contribution of each component (groundwater, soil water, and rainwater) to the surface runoff water. This is because Sakakibara et al. (2019) used average values of tracers of each component to estimate their contributions to the surface runoff water. This study assumed that rainwater did not contain ^{137}Cs and referred to the ^{137}Cs concentration in groundwater reported by Iwagami et al. (2019b). Iwagami et al. (2019b) measured the ^{137}Cs concentration in groundwater (19 samples in total) in the same catchment as this study and reported two orders of magnitude lower ^{137}Cs concentrations (0.0009 ± 0.0004 Bq/L) than in surface runoff water (Tables 2, 3). Therefore, uncertainty of the flux of rainwater and groundwater did not affect the outcome of this study. Conversely, uncertainty in the flux of soil water with a higher ^{137}Cs concentration (0.05 ± 0.03 Bq/L; Iwagami et al., 2019b; Table 2) could affect the outcome of this study.

Table 3
Surface runoff, each component contribution to the runoff water, and ^{137}Cs concentration in the runoff water during three rainfall events reported by previous studies, and the calculated ^{137}Cs discharge rate and estimated litter amount to compensate for the difference between the observed and calculated ^{137}Cs discharge rate.

		Surface runoff	Contribution to runoff water			Observed concentration in runoff water		Obs rate of discharge	Cal rate of discharge	Difference (obs-cal)	Elapsed time from rain start	Leaching rate	Estimated litter
Reference:		Sakakibara et al. (2019)				Iwagami et al. (2019b)		<This study>					
	Day, h:mm	Runoff L/s	Rain (%)	SW (%)	GW (%)	Day, h:mm	^{137}Cs (Bq/L)	$^{137}_{Obs}$Cs (Bq/s)	$^{137}_{Cal}$Cs (Bq/s)	$_{Obs}$Cs-$_{Cal}$Cs (Bq/s)	Time (hour)	Rate (Bq/g/s)	Amount (g)
Jul, 2015	16, 11:00	0.24	30.6	34.4	34.9	16, 10:30	0.031 ± 0.001	0.007	0.004	0.004	9	9.9 *10^{-6}	370
	16, 13:00	0.32	28.3	58.7	12.9	16, 13:00	0.062 ± 0.002	0.020	0.009	0.011	11	9.0 *10^{-6}	1240
	16, 15:00	0.65	6.0	27.2	66.7	16, 15:30	0.038 ± 0.001	0.024	0.009	0.016	13	8.3 *10^{-6}	1890
Aug, 2016	22, 13:00	0.14	12.6	33.7	53.7	22, 12:40	0.007 ± 0.001	0.001	Not shown because this timing is before the rain started				
	22, 15:00	0.36	13.4	36.6	50.0	22, 15:15	0.042 ± 0.001	0.015	0.006	0.009	1	27.5 *10^{-6}	320
	22, 18:00	1.03	11.3	29.7	59.1	22, 17:50	0.032 ± 0.001	0.033	0.015	0.018	4	14.4 *10^{-6}	1260
	23, 11:00	0.67	15.4	25.9	58.6	23, 11:30	0.017 ± 0.001	0.012	0.008	0.003	21	6.7 *10^{-6}	490
Aug, 2016	30, 12:00	2.28	8.0	24.9	67.1	30, 12:00	0.041 ± 0.001	0.093	0.028	0.066	39	5.0 *10^{-6}	13,180
	30, 16:00	1.80	9.7	29.9	60.4	30, 16:00	0.024 ± 0.001	0.042	0.026	0.017	43	4.8 *10^{-6}	3500
	31, 13:00	0.32	18.5	28.9	52.6	31, 12:00	0.020 ± 0.001	0.006	0.004	0.002	64	4.0 *10^{-6}	470

However, because the soil water is present in a limited space between the groundwater table and the ground surface, actual soil water contribution should not be much greater than that reported by Sakakibara et al. (2019). Even if the soil water contribution to the surface runoff water overestimates the actual flux, we consider that the overestimation would not affect the important outcome of this study. This is because overestimated soil water fluxes lead to lower estimates of dissolved ^{137}Cs concentrations in surface runoff water.

4.3. Saturated surface area expansion during rainstorms

The relationship between surface runoff rate and estimated saturated surface area is shown in Fig. 6. The surface runoff rate in the 11 tested cases was between 0.06 and 1.12 L/s. In the cases where the surface runoff rate was higher than 1.12 L/s, the saturated surface area was larger than the shooting range of the interval camera. Therefore, we could not estimate this area. The estimated saturated area in the tested cases was between 0.06 and 25.26 m^2. The dotted line in Fig. 6 represents the regression curve of the surface runoff rate and saturated surface area. It is evident that the saturated surface area expands exponentially as the surface runoff rate increases.

4.4. Discharge of dissolved ^{137}Cs during rainstorms

The data from the three rainfall events (Table 3) are shown in Fig. 7. These data include rainfall, surface runoff rate, contribution of endmembers to the surface runoff water, observed ^{137}Cs discharge rate, and calculated ^{137}Cs discharge rate with consideration given to hydrological processes. The observed ^{137}Cs discharge rate increased with increasing surface runoff rates (Iwagami et al., 2019b). Additionally, there was a hysteresis in the ^{137}Cs discharge rate; the ^{137}Cs discharge decreased at a slower rate than the rate at which the ^{137}Cs discharge increased. Differences between the observed and calculated ^{137}Cs discharge rates were more significant at high than at low surface runoff rates.

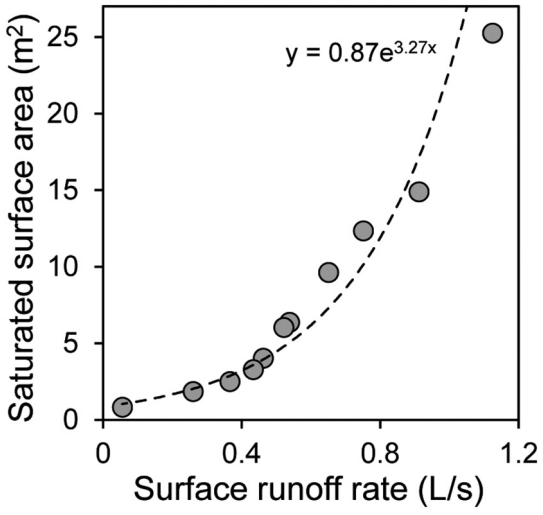

Fig. 6. Relationship between surface runoff rate and the estimated saturated surface area.

Previous research has indicated that factors such as soil water, throughfall, and litter accumulated on the ground surface contribute to elevated levels of dissolved ^{137}Cs discharge (e.g., Tsuji et al., 2016; Iwagami et al., 2019b). To improve the indication, this study examined the calculation of ^{137}Cs discharge rate during rainstorms without considering ^{137}Cs leaching from litters and only with the consideration of endmember mixing, which includes soil water, groundwater, and rainwater components (Fig. 7). The calculations underestimated the observed values. This suggests another factor (the leached ^{137}Cs from litter) contributing to elevated levels of dissolved ^{137}Cs in the surface runoff water during rainstorms in the headwater.

To discuss the mechanism of the elevated dissolved ^{137}Cs concentration in the surface runoff water during rainstorms, we focused on the expansion of the saturated surface area. The saturated surface area exponentially expanded during rainstorms (Fig. 6) owing to the rise in the groundwater table level (Iwagami et al., 2019b). Subsequently, the area started to flood as a saturated overland flow. This overland flow contributed to the mainstream as a new surface runoff contribution only during rainstorms.

Conversely, the relationship between the surface runoff rate at the analyzed time and the estimated litter amount that compensates for the difference between the observed and calculated ^{137}Cs discharge (Table 3) is presented in Fig. 8. The dotted line in Fig. 8 represents the regression curve obtained using an exponential approximation. The trend seems to be that the estimated litter amount increases with increasing surface runoff rate. This trend is consistent with that of the saturation surface area expansion (Fig. 6). This indicates that the saturated surface area exponentially expands as the groundwater table level rises during rainstorms, and the contact area between the water and litter on the ground surface also exponentially expands. This could induce acceleration in ^{137}Cs leaching.

To examine the validity of the estimated amount of litter (Fig. 8), the litter density (g/m^2: amount per unit area) was calculated using the regression equation in Fig. 6 and the observed surface runoff rate (x-axis data in Fig. 8). Since the data with a surface runoff rate of more than 1.2 L/s is out of the range of the regression equation, we excluded two data in Fig. 8 from the calculation. The calculated litter density was between 50 and 500 g/m^2 (average ± SD: 200 ± 150 g/m^2). Assuming

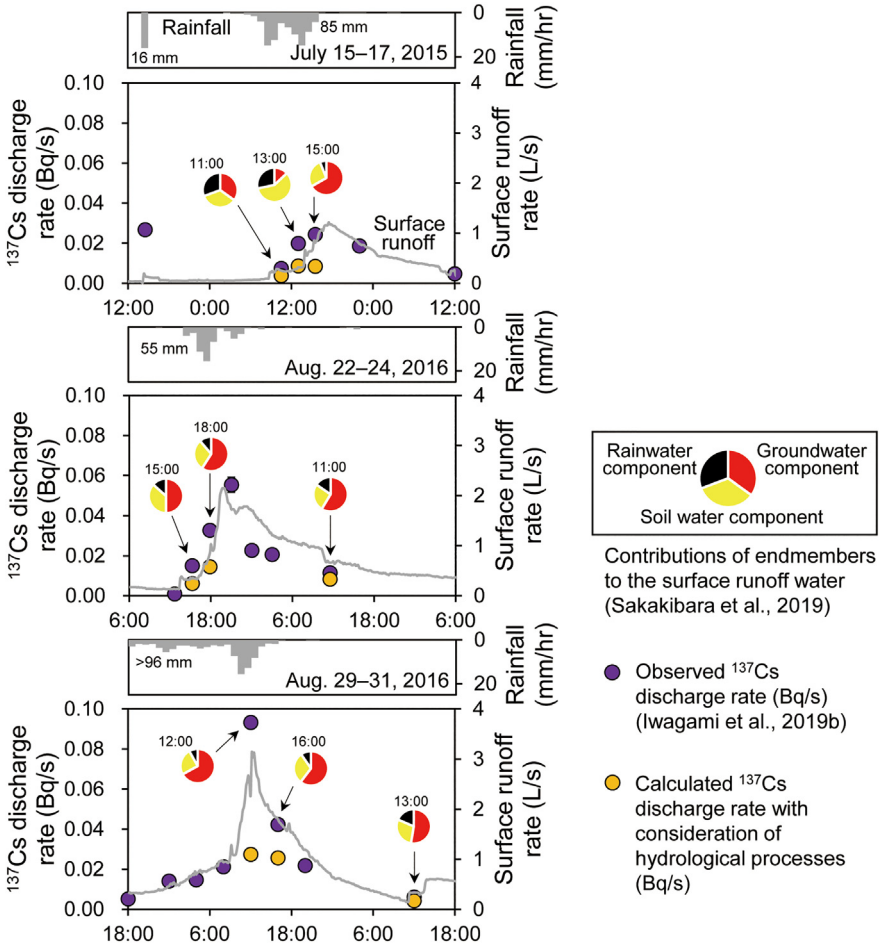

Fig. 7. Rainfall, surface runoff, contributions of endmembers to the surface runoff water, observed ^{137}Cs discharge rate, and calculated ^{137}Cs discharge rate with consideration of hydrological processes in the three rainfall events shown in Table 3.

that the weight of each leaf litter is 1 g, there will be an average of 200 leaves per square meter. Although actual litter density was not observed, this value is not far from reality. Hence, the cause of elevated dissolved ^{137}Cs in the surface runoff water could be explained in terms of

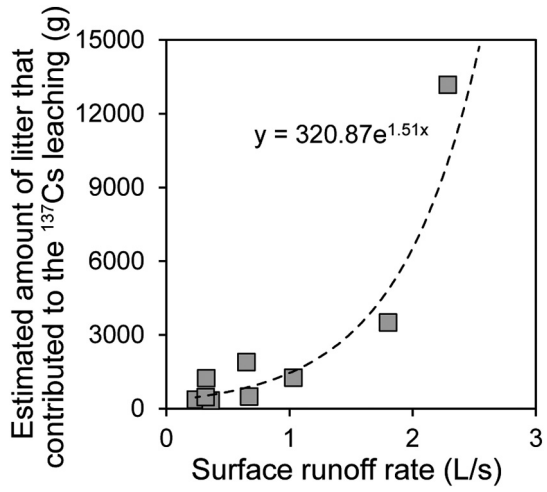

Fig. 8. Relationship between surface runoff rate at the analyzed time and the estimated litter amount for compensating the difference between observed and calculated ^{137}Cs discharge rate.

^{137}Cs leaching due to the expansion of the saturated surface area. However, large variation in the calculated litter density and different shapes of the exponential curves in Figs. 6 and 8 may indicate that there is uncertainty in the assumptions made in this study. This uncertainty is discussed in this section.

Presence of contaminated litter and conditions that produce saturated surface flows are considered principal factors that contribute to the ongoing release of elevated dissolved ^{137}Cs during rainstorms in FDNPP-accident-affected forested headwaters. This interpretation is supported by Teramage et al. (2014), who found that litter on the forest floor, laterally transported from a hillslope (Koarashi et al., 2014), contains a high ^{137}Cs concentration.

Similar leaching results of both agitation and non-agitation cases in this study (Fig. 4) seem to strengthen this interpretation. Saturated surface areas, which are formed by rising groundwater table level during rainfall, are new source areas of surface runoff. However, these areas did not generate strong surface runoff. In other words, test conditions without agitation may be more realistic. Then, after outflowing and becoming a stream, the stream water and litter on the ground would be better reflected in the agitating experiments. The fact that ^{137}Cs leaching occurred in both agitation and non-agitation cases is important, and therefore additional leaching of ^{137}Cs from litter would occur in all processes generating saturated overland flow during rainstorms.

Increasing ^{137}Cs leached from litter due to the expansion of the saturated surface area is expected to occur in most rainfall events that the saturated surface flow generates. This is because the dilution of dissolved ^{137}Cs due to rainfall input (Nagao et al., 2013) may not occur in

forested headwaters for the following two reasons. First, regardless of the rainfall intensity, contribution of rainwater to direct runoff is limited due to the high porosity and permeability of forest soil. Second, no matter how short the contact time between the litter and the water is, leaching of ^{137}Cs from the litter may occur. Sakuma et al. (2021) explained that a large amount of ^{137}Cs leaches from litters immediately upon soaking in water because of the existing weak-bonding water-extractable ^{137}Cs. However, there should be an upper limit to the expansion of the saturated surface area (estimated to be approximately 80 m^2 in this study). In other words, if the rainfall exceeds the amount of rainfall when the maximum saturated surface area is formed, the contact area between water and litter will not increase any further. When such a situation occurs, the dissolved ^{137}Cs concentration in surface runoff water in forested headwaters may start to decrease because the surface runoff water is mainly composed of groundwater with low dissolved ^{137}Cs concentrations (Iwagami et al., 2019b; Sakakibara et al., 2019).

Kurihara et al. (2020) reported that ^{137}Cs was leached from the decomposed litter, although the leaching rate of ^{137}Cs was lower than that of fresh litter. Therefore, it is necessary to consider ^{137}Cs leaching from the decomposed litter layer in this study. However, this study did not collect decomposed litter and did not include it for the calculation. This is because the dissolved ^{137}Cs in soil water reported by Iwagami et al. (2019b) was considered in our mixing analysis. Iwagami et al. (2019b) collected soil water samples from the same valley as our study (Fig. 1), and targeted soil water included the water in the decomposed litter layer. Considering rainfall-runoff processes in the studied catchment, surface runoff was generated as a result of the mixing of rainwater, soil water, and groundwater (Sakakibara et al., 2019). Soil water is water that has already been stored in the subsurface area, including the area close to the ground surface, before rainfall starts. Soil water is always present in the valley of the study area; hence, soil water should contain dissolved ^{137}Cs leached from decomposed litter in addition to soil particles. Therefore, we considered it unlikely that the decomposed litter layer would be an important new source of dissolved ^{137}Cs during rainstorms. However, no studies have clarified the temporal variations in dissolved ^{137}Cs concentrations in soil water during a single rainstorm event. Therefore, we could not be certain that there would be no new leaching from decomposed litter in forested headwaters. This point should be examined in future studies.

Another uncertainty in this study is related to the fact that it did not consider the degree of ^{137}Cs leaching and degradation process of the litter from litter falls to rainstorm events. This is because the timing of the litter sampling (November 2015) and the targeted rainfall events (July 2015 and August 2016) were very different. Since ^{137}Cs concentration in the litter was much higher than that in environmental water (Section 4.1 in this study; Iwagami et al., 2019b; Teramage et al., 2014), the litter would likely retain a high concentration of ^{137}Cs even if some ^{137}Cs leached out. However, previous studies have reported that ^{137}Cs leaching rate decreases when the litter is decomposed by dry and wet processes and temperature changes (Kurihara et al., 2020; Sakuma et al., 2021). If this is true, ^{137}Cs leaching rate (μBq/g/s) from the litter (Fig. 5) would be lower, and the estimated litter quantity (Fig. 8; Table 3) would be larger. Thus, room for discussing specific quantity exists, but this uncertainty increases the difference between the observed and calculated dissolved ^{137}Cs discharge rates. Therefore, the main interpretation of this study is that the ^{137}Cs leaching from litter deposited on the ground surface is accelerated with expansion of the saturated surface area, supported by the results of this study.

According to Sakai et al. (2015), Kurihara et al. (2020), and this study (Fig. 4), a large amount of ^{137}Cs activity remained in the litter samples after leaching tests, although the percentage varied among studies. Considering that the leaching rate after the test remained low, the litter would prolong the maintenance and discharge of the FDNPP accident-oriented ^{137}Cs in the forest. Nevertheless, even if the ^{137}Cs leaching occurs from the litter, it can continue to act as a new additional ^{137}Cs source contributing to elevated dissolved ^{137}Cs in the surface runoff water during rainstorms. Sakai et al. (2015) referred to Sheppard and Evenden (1990) and mentioned that "radiocesium leaching from the litter can occur for up to 300 days." However, few studies have tested ^{137}Cs leaching from litter using FDNPP accident-affected samples for more than 30 days. For a more detailed discussion about the prolonged influence of dissolved ^{137}Cs discharge from the forest, a longer timescale test may be necessary.

Finally, regional characteristics of dissolved ^{137}Cs concentrations in surface runoff water during rainstorms are discussed. Several years after the FDNPP accident, increase in dissolved ^{137}Cs concentration in the surface runoff water during rainstorms was larger in the contaminated and forested headwater (catchment area: less than 0.05 km^2; Iwagami et al., 2019b) than in the downstream near the forest (catchment area: 21 km^2; Tsuji et al., 2016). This difference might be explained by ^{137}Cs leaching from litter, which is a new ^{137}Cs source, caused by the expansion of the saturated surface area during rainstorms in the forested headwater. Conversely, in the downstream area of the forest, although ^{137}Cs originating from the litter at the headwater should be loaded to the surface runoff water, the concentration may be diluted by water with a lower ^{137}Cs concentration, such as discharge at the lower ^{137}Cs inventory area and at litter-free areas (e.g., concrete roads and paddy fields). This interpretation can extend to the result of Sakuma et al. (2019), who found that the K_d absorption/desorption model without considering ^{137}Cs leaching from organic matter could not reconstruct dissolved ^{137}Cs concentrations in the surface runoff water during rainstorms. This study found a relationship between the quantity of ^{137}Cs leached from litter and the saturated surface area associated with hydrological processes during rainstorms in forested headwaters. This suggests that it is necessary to consider not only the leaching rate of ^{137}Cs from litter but also the spatiotemporal variations of the saturated surface area to improve the model regarding the dissolved ^{137}Cs discharge from forests.

5. Conclusions

The leaching characteristics of ^{137}Cs from broadleaf litter were examined using laboratory leaching tests. Processes of elevated levels of dissolved ^{137}Cs in the surface runoff water during rainstorms were discussed in terms of ^{137}Cs leaching from litter and hydrological processes. The findings of this study are as follows:

1. Leaching test results indicated that the leaching rate was higher during the first hour, indicating immediate ^{137}Cs leaching after the start of soaking; therefore, the elapsed time from the start of the rainfall is important when the ^{137}Cs leaching during a rainstorm is considered.
2. Leaching tests were conducted with agitation (100 rpm) and non-agitation (0 rpm) based on the situated consideration of saturated overland flow generation in the headwaters. The results showed no significant difference between them, indicating that ^{137}Cs leaching from litter would occur in processes generating saturated overland flow during rainstorms.
3. Leaching rate of ^{137}Cs from litter samples after the 24-h test was up to 33.7%, indicating that a large part of the original ^{137}Cs activity remained in the litter samples. This result suggests that the litter would prolong the maintenance and discharge of the FDNPP accident-oriented ^{137}Cs in the forest, as previously mentioned.
4. Dissolved ^{137}Cs concentrations in the surface runoff water in the headwater during rainstorms could not be reproduced by considering mixing among endmembers (rainwater, soil water, and groundwater). Differences between observed and reproduced ^{137}Cs in the surface runoff water were larger under high-flow conditions. This indicates an additional ^{137}Cs loading factor that impacts surface runoff water during rainstorms.

5. Saturated surface area expanded as the surface runoff rate increased in the headwaters. This indicates that the contact area between surface runoff and litter accumulated on the forest floor increased.
6. Leaching of ^{137}Cs from the litter in the saturated surface area that temporarily forms during rainstorms could be a cause of the elevated dissolved ^{137}Cs in the surface runoff water from rainfall-runoff events in the headwater. However, this study did not consider ^{137}Cs leaching from the decomposed litter layer, degradation processes of litter during the study period, and extreme rainstorm cases. These are the sources of uncertainty; therefore, further research addressing these issues is necessary to validate the findings of this research.
7. The results and interpretation of this study could be applied to any forested catchments that satisfy two conditions: contaminated litter is present, and saturated surface flows are generated during rainstorms. Therefore, these conditions are suggested as principal factors that contribute to the ongoing release of elevated dissolved ^{137}Cs during rainstorms in FDNPP-accident-affected forested headwaters.
8. This study considered the relationship between ^{137}Cs leaching from litter and expansion of the saturated surface area during rainstorms in forested headwaters. Because of this relationship, to improve the model for evaluating the dissolved ^{137}Cs discharge from forests, it is necessary to include not only the ^{137}Cs leaching characteristics from the litter but also the spatiotemporal variations of saturated surface flow generation associated with hydrological processes.

CRediT authorship contribution statement

Koichi Sakakibara: Writing – original draft, Formal analysis, Data curation, Visualization. **Sho Iwagami:** Writing – review & editing, Methodology, Validation. **Maki Tsujimura:** Conceptualization, Project administration. **Ryohei Konuma:** Investigation. **Yutaro Sato:** Investigation. **Yuichi Onda:** Conceptualization, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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