

Differences in leaching characteristics of dissolved radiocaesium and potassium from the litter layer of Japanese cedar and broadleaf forests in Fukushima, Japan

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ABSTRACT

Cesium is an element that belongs to the same group as K, and is known to show similar behaviour to that of K in plants. In this study, we conducted a serial leaching test for 120 h to compare the leaching characteristics of dissolved ^{137}Cs and K in forest litter, obtained from Japanese cedar and deciduous broadleaf forests located 40 km from the site of the 2011 Fukushima Dai-ichi nuclear power plant accident. The litter was collected in 2018 and was divided into three groups according to the decomposition level. The cumulative leachable fraction of ^{137}Cs at 120 h ranged from 0.3% to 3.3%, suggesting that most of the ^{137}Cs in the litter was hardly leachable in water. The leachable fraction of ^{137}Cs generally decreased with the decomposition level of the litter, implying that the easily leachable ^{137}Cs eluted into the water during the first stage of decomposition. Meanwhile, the cumulative leachable fraction of K at 120 h was approximately 10 times greater than that of ^{137}Cs and ranged from 22.7% to 54.8%. The leaching speeds of ^{137}Cs and K decreased suddenly with elapsed time regardless of the tree species, decomposition degree, or element. Our findings contribute to the long-term understanding of the ^{137}Cs cycle in forest ecosystems.

1. Introduction

Radioactive contamination in forest areas following a nuclear accident is difficult to control using techniques, such as decontamination, compared with other ecosystems such as an agricultural land and an urban area. Studies conducted after the Chernobyl nuclear power plant accident in 1986 have shown the prolonged contamination of forest ecosystems and products (IAEA, 2010). Food monitoring for radioactive substances after the Fukushima Dai-ichi nuclear power plant accident in 2011 have also shown that wild foods from forests still have higher concentrations of ^{137}Cs than agricultural crops (Uekusa et al., 2015). An appropriate understanding of the migration of ^{137}Cs in the forest ecosystem is required for the long-term forest management, resumption of forest industry and utilisation, and radiation protection of forest users after a nuclear accident.

On a long-term basis, forest litter is one of the most important components of the ^{137}Cs cycle in a forest ecosystem. Cesium-137 deposited on a forest floor shortly after a nuclear accident was observed to migrate into the soil for a while through litter decomposition, and the fraction absorbed by plants was returned to the forest floor as litterfall (Shaw, 2007). As ^{137}Cs leached from the litter layer mainly exists in a dissolved form (Takada et al., 2016) that is highly

bioavailable (Nakanishi et al., 2013), a portion of ^{137}Cs continues to circulate in the forest ecosystem (Goor and Thiry, 2004). Therefore, contribution of the litter layer to the ^{137}Cs cycle in a forest ecosystem needs to be examined thoroughly.

Several studies have been conducted focusing on the migration of ^{137}Cs from litter using litterbags and lysimeters (Rafferty et al., 1997; Huang et al., 2016; Kurihara et al., 2018a, 2018b; Nakanishi et al., 2014; Koarashi et al., 2016). These studies were conducted under actual environmental conditions and quantitative results for downward migration of ^{137}Cs were obtained; however, the information obtained is very site-specific and does not particularly address the leaching mechanism. Some leaching tests have been conducted in laboratories, however most tests were adapted to the natural conditions similar to a field experiment. For example, Sakai et al. (2015) conducted a leaching test in laboratory to examine the adsorption of ^{137}Cs and K on soil particles in river water, Saito et al. (2015) studied the leaching of ^{137}Cs from well-decomposed litter (FH layer) in addition to the soil layers by conducting a leaching test, and Saito et al. (2017) adopted leaching tests to litter-bags installed in an actual environment. These studies did not evaluate the fundamental leaching characteristics such as leaching capacity and the relation with litter decomposition, and the mechanisms were not examined. The laboratory experiments focusing on the

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fundamental leaching characteristics will provide an important knowledge.

Potassium is an element that belongs to the same group as Cs and is known to show similar behaviour in plants (Kruglov et al., 2005). Like K, plants absorb Cs via roots (Broadley and Willey, 1997), and ^{137}Cs shows almost the same distribution as K among all the tree components (Goor and Thiry, 2004). While studies on the similarity of K and Cs in a plant have been conducted, knowledge on the similarity of leaching characteristics of K and Cs eluted from a dead plant (e.g., litter) is still insufficient. As K has long been known to leach easily from litter (Lousier and Parkinson, 1978; Osono and Takeda, 2004), several studies indicated that ^{137}Cs in litter does not have similar properties as K. Saito et al. (2015, 2017) reported that ^{137}Cs is difficult to elute from litter by conducting a leaching test; however, they did not evaluate K. Sakai et al. (2015) also observed that the concentrations of both ^{137}Cs and K in litter decreased by putting litter in water, and found that the decrease in K was greater than that of ^{137}Cs . A litterbag experiment after the Chernobyl accident also showed that the decrease of K in the litter was faster than that of ^{137}Cs (Rafferty et al., 1997). Knowledge on the similarity or non-similarity between the leaching characteristics and mechanisms of ^{137}Cs and K in litter can support the long-term prospect of ^{137}Cs dynamics in a forest ecosystem.

In this study, we conducted a serial leaching test with reference to the tank leaching test (NEN 7345, 1994; Malviya and Chaudhary, 2006; Sakanakura et al., 2003) to examine the leaching characteristic of dissolved ^{137}Cs from litter. We also compared the leaching characteristics of ^{137}Cs and K.

2. Materials and methods

2.1. Litter samples

The litter samples for the leaching test were collected from two types of typical forest in Kawamata Town, Yamakiya District, located approximately 40 km northwest of the Fukushima Dai-ichi nuclear power plant (FDNPP): a Japanese cedar (*Cryptomeria japonica*) forest (37°35' N, 140°41' E; 550 m asl) and a deciduous broadleaf forest (37°36' N, 140°40' E; 580 m asl), dominated by konara oak (*Quercus serrata*). The total atmospheric deposition of ^{137}Cs from the FDNPP accident was estimated to be 470 kBq/m² and 440 kBq/m² in the Japanese cedar and deciduous broadleaf forests, respectively, by the Third Airborne Monitoring Survey of radioactive contamination (Ministry of Education, Culture, Sports, Science and Technology, 2011).

In each forest, two or three layers of litter (L1, L2, and F layers) were collected separately to evaluate the effect of litter decomposition on leaching characteristics. L1 consisted of recently fallen leaves, L2 contained relatively old but unfragmented leaves, and F contained fragmented organic materials (Fig. 1). Sample collection was conducted on August 28, 2018. Because L1 was absent from the forest floor in the deciduous broadleaf forest in August, the litter samples from this forest were only L2 and F layers. All samples were stored in a refrigerator at 4 °C until the implementation of the leaching test on 19–24 September 2018. For the litter samples in the present study, ^{137}Cs existing in plant tissues was assumed to be predominant, the lifetime of cedar needles at crown is reported to be approximately 3 years (Sugawara, 1970), and the litter turnover of cedar is reported to be from 3 years to 5 years (Ichikawa et al., 2003). As the turnover of litter layer in deciduous broadleaf in west Japan is reported to be 1.5 years (Rahman and Tsukamoto, 2013), the decomposition of litter in which ^{137}Cs deposited at the time of the FDNPP accident was completed until the sampling.

Each litter sample was mixed homogeneously, and 40–60 g of wet litter samples was dried at 95 °C for 24 h, and thereafter crushed. The crushed litter was placed in plastic containers (100 ml of diameter 5 cm), and ^{137}Cs activities were measured using a high-purity germanium (HPGe) detecting system (Canberra-Eurisys). The measured time was 120,000–200,000 s, depending on the radioactivity of the samples.

Cedar



Broadleaf



Fig. 1. Litter layers from cedar forest (above) and broadleaf forest (below) used for the leaching test. (Left to right) L1, recently fallen leaves; L2, relatively old but unfragmented leaves; F, fragmented organic material.

Efficiency calibration of the detector used for the measurements was performed using two types of mixed radionuclide reference standard sources (Isotope Products Laboratories No.752-69, Eckert & Ziegler Isotope Products No. 1390-63). The counting errors were usually lower than 10%. The total K content in the litter sample (mg/kg-dry) was determined by atomic absorption spectrometry after microwave digestion.

2.2. Leaching test and measurements

A leaching test was performed based on the tank leaching test (NEN 7345, 1994; Malviya and Chaudhary, 2006; Sakanakura et al., 2003) for five types of litter samples. The litter samples for the experiment were wet as they were in a natural condition and were not subjected to drying treatment. The weight of each wet litter sample was 100–180 g, equivalent to 46 g dry weight. Samples were placed in deionized water in polypropylene beakers and mixed using a magnet stirrer. Repetition of the test was one for each type of litter. The solid/liquid ratio was 1:50. There is a possibility that the leachable fraction is affected by litter degradation taking place during a long-term leaching test. We only focused on leaching immediately after water contact in the present study. Therefore, the beakers were kept at 4 °C to avoid litter degradation during the leaching test and the leaching test was completed within 5 days. The leachate was separated six times after 0.5, 7.5, 15, 30, 60, and 120 h during the 5-day (120 h) leaching test, with new leaching solution added each time. Accordingly, 30 samples were produced in total. The solutions from the leaching test were filtered using 0.45 µm-pore-sized mixed cellulose ester membrane filters to remove suspended solids.

After an aliquot of the solutions was separated for the quantification of K ions, the remaining was evaporated to approximately 5 ml for the radioactivity measurement of ^{137}Cs . Small amounts of organic matter precipitated during the evaporation process. The concentrations of ^{137}Cs

activity in the evaporated solutions were measured using a well-type HPGe detecting system (Canberra-Eurisys). The measured time was 1223–251,040 s, depending on the radioactivity of the samples until the peak counting error was below 10%. Efficiency calibration of the detector used for the measurements was performed using the mixed radionuclide standard reference material IAEA-444. The detection limit was 2.2–74.2 Bq/kg. The leachable fraction was calculated from the activity concentrations of solutions and litter. The activities of ^{137}Cs in litter and solutions were corrected for radioactive decay to March 11, 2011. The concentration of K in the solutions were measured using an inductively coupled plasma optical emission spectrometer. The detection limit was 0.05 mg/L. The leachable fraction of K in the litter was calculated in the same way as ^{137}Cs .

3. Results

3.1. Properties of litter samples

Table 1 shows the activity concentrations of ^{137}Cs (kBq/kg-dry) and K content (mg/kg-dry) of the litter samples. The C/N ratio and the loss on ignition of the litter samples collected in December 2017 are also shown as a reference. The criterion of the litter layers (L1, L2, and F layers) is same regardless of the sampling season. The activity concentrations of ^{137}Cs range from 0.6 to 57.4 kBq/kg, indicating that the activity concentration increases with the degree of litter decomposition in both cedar and broadleaf forests, as seen in previous studies (Huang et al., 2016; Ichikawa et al., 2015). The K content increases with the degree of decomposition, but the trend is less apparent compared to ^{137}Cs . The C/N ratio of litter in December 2017 ranged between 22.9 and 67.2 and decreased from L1 to L2 to F. The C/N ratio of litter is known to decrease with the degree of litter decomposition (Osono and Takeda, 2001), and the same trend was observed in the present study. Loss on ignition (%) of litter in December 2017 was lower in the F layer than in the L1 and L2 layers.

3.2. Temporal changes in the cumulative leachable fraction of ^{137}Cs and K

The concentrations of ^{137}Cs (Bq/L) and K (mg/L) in the leachates from each litter layer are shown in Table S1. The activity concentrations of ^{137}Cs ranged from 0.02 Bq/L to 2.8 Bq/L. The highest concentration was observed in the F layer of the broadleaf litter at 0–0.5 h, whereas the lowest concentration was in the L1 layer of the cedar litter at 7.5–15 h. The concentrations of K in the solutions ranged from 0.5 to 3.2 mg/L. The highest was in the L2 layer of the broadleaf litter at 0–0.5 h, whereas the lowest was in the L1 layer of the cedar litter at 0–0.5 h and 7.5–15 h.

A clear trend in the activity concentrations of ^{137}Cs in the solutions for the L1 cedar litter layer, according to an increase in the leaching time, was not observed, however the concentration was the highest during the final 60–120 h. Potassium also showed a similar trend as

Table 1

Properties of litter samples collected on August 28, 2018 for the tank leaching test.

Forest type	Layer*	^{137}Cs activity concentration (kBq/kg-dry)**	K content (mg/kg-dry)	C/N ratio***	Loss on ignition*** (%)
Cedar	L1	0.606	330	67.2	96.3
	L2	9.28	500	45.3	95.4
	F	57.4	1180	22.9	86.2
Broadleaf	L2	9.38	970	29.4	93.6
	F	27.2	1370	27.7	85.9

*L1, recently fallen leaves; L2, relatively old but unfragmented leaves; F, fragmented organic material.

** Counting error is under 0.6% of activity concentration.

*** Values for samples collected on December 27, 2017.

^{137}Cs according to the degree of litter decomposition and leaching repetition.

Fig. 2 depicts the cumulative leachable fraction of ^{137}Cs and K over time from each type of litter material. The leaching of ^{137}Cs and K rapidly decreased with leaching time. The leachable fraction of ^{137}Cs at 120 h ranged from 0.3 to 3.3%, with the L2 broadleaf litter layer exhibiting the highest value, and the L2 and F layers of the cedar litter showing the lowest values (Table 2). For K, the leachable fraction at 120 h was approximately 10 times higher than ^{137}Cs at 22.7–54.8%. The highest value was obtained from the L1 layer of the cedar litter, and the lowest was from the F cedar layer. We tried to adopt the dual exponential model for understanding the change in leachable fractions over time by using a least square fitting method, but almost half of regression coefficients were not statistically significant.

3.3. Temporal changes in the leaching speed of ^{137}Cs and K

Temporal changes in the leaching speed with respect to the leaching time are shown in Fig. 3. Both for ^{137}Cs and K, the log-transformed leaching speed linearly decreased with the log-transformed leaching time. The leaching speeds of all the data decreased 2–3 orders of magnitude over 100 h, regardless of the degree of litter decomposition, species of trees, and elements. NEN 7345 (1994) and Sakanakura et al. (2003) suggest that a change in the gradient during the leaching indicates that the leaching mechanism changed during the test. Fig. 3 shows that no change in the gradient was observed over 5 days. Therefore, we concluded that only one type of leaching mechanism was dominant during the test.

4. Discussion

This study evaluates the leachable fraction of ^{137}Cs and K in litter by conducting a leaching test in a laboratory. The leaching speed of ^{137}Cs and K was 1/100–1/1000 of the initial state after 120 h of the leaching test (Fig. 3), and it is unlikely that leaching would significantly increase beyond this time, suggesting that ^{137}Cs was mostly leached in the present study. However, additional tests are required to discuss leachable fraction of ^{137}Cs and K over 120 h and the leachable capacity.

Some studies have been conducted on radiocesium leaching from litter as shown in Table 2. Sakai et al. (2015) reported that the concentration of ^{137}Cs in fresh cedar litter decreased by approximately 30% when the samples were soaked for 1 day at 21 °C. Their result showed a significantly larger leachable fraction than our result for the L1 cedar layer after 120 h. The higher temperature during leaching may have enhanced leaching due to litter degradation during the leaching test. For broadleaf litter, observed a lower (Saito et al., 2015) or a higher (Saito et al., 2017) leachable fraction than our results. This is because they adopted a different experimental condition; they used more decomposed litter materials from the H layer or the contents of the litter-bag composed of fresh litter, and they dried the litter well at 80 °C before performing the leaching test. The objectives of these previous studies were not to understand the fundamental leaching characteristics such as leaching capacity or relation to litter decomposition. In addition, as the experimental conditions such as temperature and wet/dry condition of the litter materials differed from our study, a simple comparison between our study and previous studies is not appropriate. The data of our study is crucial information regarding the fundamental leaching characteristics of ^{137}Cs from litter.

The measured leachable fraction of ^{137}Cs at 120 h (Table 2) from broadleaf litter was higher than that from cedar litter, demonstrating that ^{137}Cs from the broadleaf litter was easier to elute into water compared with that in the cedar litter. A field experiment conducted using a lysimeter in Fukushima showed that the migration rate of dissolved ^{137}Cs from the litter layer of a broadleaf forest was more than three times higher than that from a cedar forest (Kurihara et al., 2018b); our results support the findings of this field experiment.

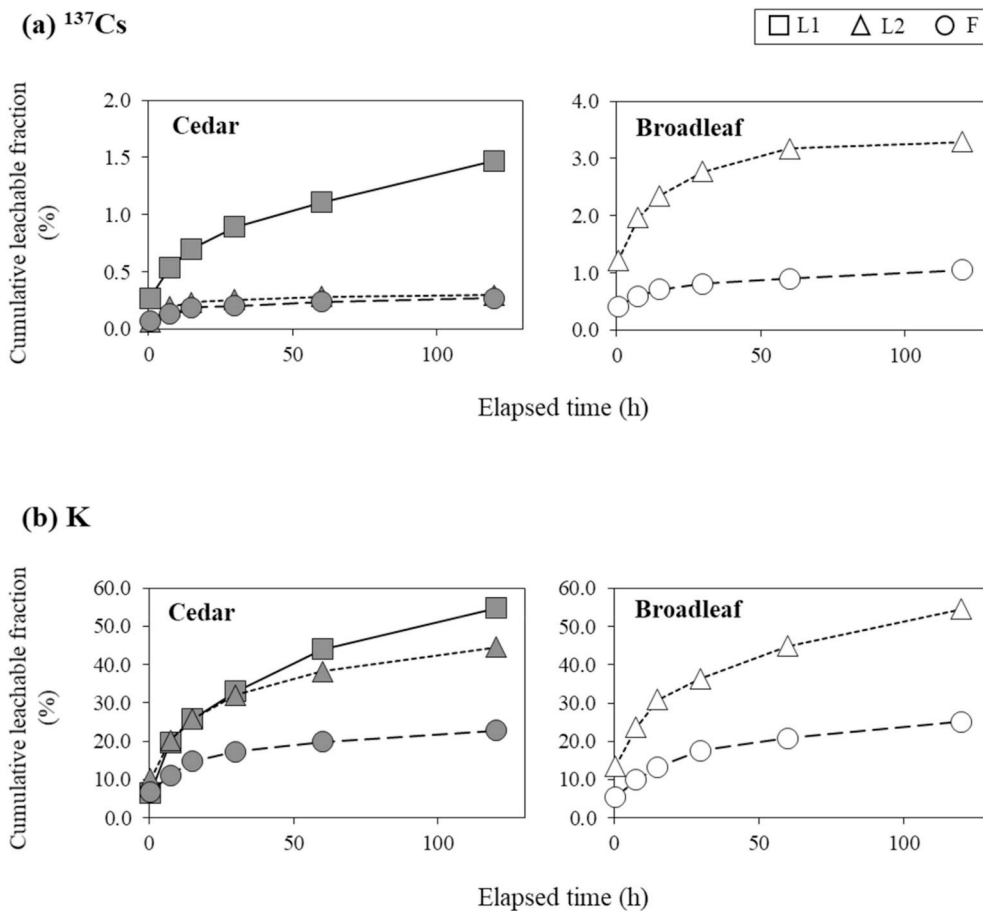


Fig. 2. Cumulative leachable fraction of ^{137}Cs and K over time from each litter layer (L1, L2, and F) in cedar and broadleaf forests.

Table 2

Leachable fraction of ^{137}Cs and K at 120 h in the present study and a comparison with the results of previous studies.

Litter type	Leachable fraction (%)		Source
	^{137}Cs	K	
Cedar L1	1.5	54.8	This study
Cedar L2	0.3	44.5	This study
Cedar F	0.3	22.7	This study
Broadleaf L2	3.3	54.5	This study
Broadleaf F	1.1	25.1	This study
Fresh cedar litter	34	89	Sakai et al. (2015): 24 h of leaching at 21 °C.
Broadleaf FH	0.03–0.04	–	Saito et al. (2015): Dried at 80 °C for 120 h, 6 h of leaching at room temperature.
Broadleaf litter bag	Approximately 10–40	–	Saito et al. (2017): Dried at 80 °C for 48 h, 30 min of leaching.

The leachable fraction at 120 h generally decreased with litter decomposition, indicating that easily leachable ^{137}Cs leached into the water first. The leachable fraction of ^{137}Cs in litter is thought to have decreased with litter decomposition (i.e., from L1 to F). This trend was also observed in a field experiment by a previous study; Saito et al. (2017) conducted a laboratory leaching test with litter bags placed in the broadleaf forest in Fukushima and found that the leachable fraction of ^{137}Cs in water decreased with litter decomposition. However, for the L2 and F layers of the cedar litter, similar temporal changes in the

cumulative leachable fraction were observed in Fig. 2, although there are significant differences between them with respect to the fragment size of the litter and the C/N ratio, as shown in Fig. 1 and Table 1.

The leachable fraction of K decreased with the degree of litter decomposition in the present study, as was also observed for ^{137}Cs . Our result in the laboratory suggests that the leaching potential of ^{137}Cs is significantly lower than that of K which supports several results of previous studies. For example, a rapid decrease in K in pine litter compared with ^{137}Cs was found in a litterbag experiment conducted in Chernobyl and Ireland (Rafferty et al., 1997).

The lower leachable fraction of ^{137}Cs at 120 h than that of K implies a higher abundance ratio of the hardly leachable form of ^{137}Cs in litter compared with that of K. Many studies revealed that Cs is one of the competitors of K in terms of root uptake and transportation in plant bodies (e.g., Kruglov et al., 2005; Broadley and Willey, 1997). Meanwhile, the measured leachable fractions of ^{137}Cs in the litter samples after 120 h obtained in the present study were 1–2 orders of magnitude smaller than those of K. These results suggest ^{137}Cs and K are present in the litter in different chemical forms. However, it is not clear whether the chemical forms of ^{137}Cs and K are already different prior to defoliation, or if they are altered by environmental conditions after defoliation. Further studies are required on the existing chemical form of ^{137}Cs in plants which will help to closely understand the leaching mechanisms of ^{137}Cs from litter, and consequently, the ^{137}Cs cycle in forest ecosystems.

5. Conclusions

The present study focused on the contribution of the litter layer on the ^{137}Cs cycle in two forest ecosystems. This is the first study to examine

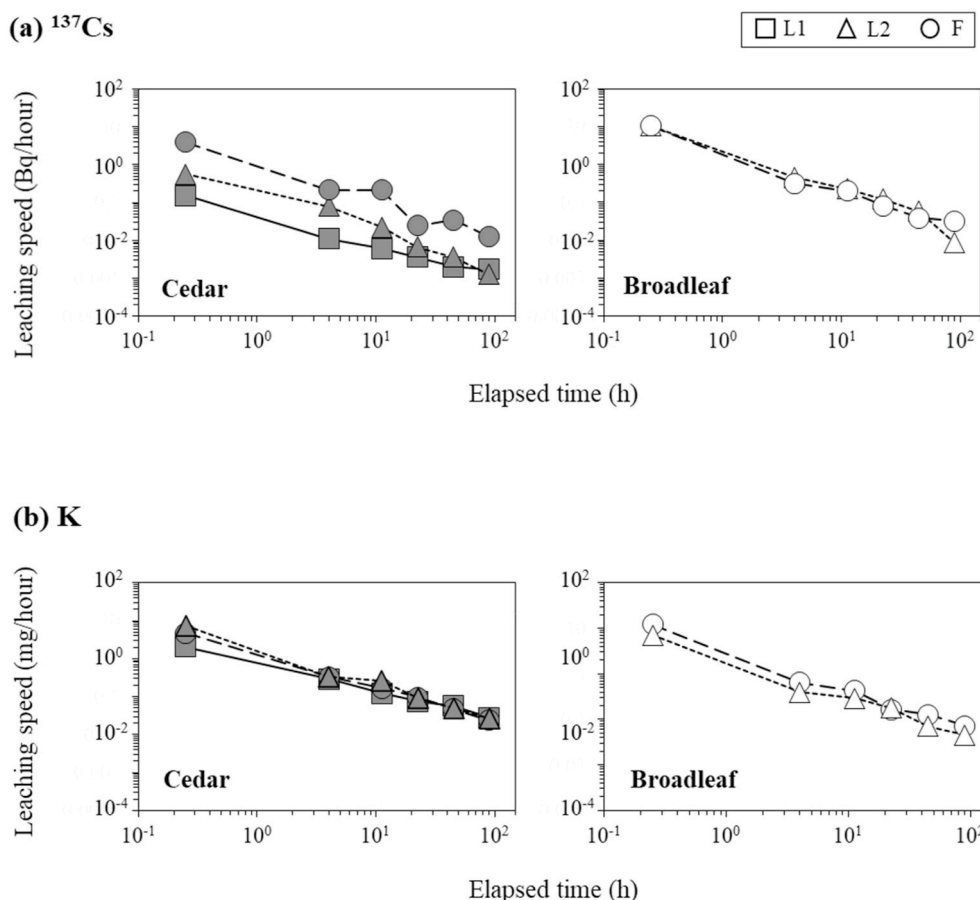


Fig. 3. Temporal change in ^{137}Cs and K leaching per unit time from each litter layer (L1, L2, and F) in broadleaf and cedar forests.

the fundamental leaching characteristics, i.e. the leachable fraction of ^{137}Cs and K in litter and their relation to the degree of litter decomposition. The cumulative leachable fraction of ^{137}Cs at 120 h was 0.3–3.3%, indicating that most ^{137}Cs in litter was hardly leachable in water. This trend was particularly strong for the well-decomposed litter. The leachable fraction from the F layer (fragmented organic material) was lower than that from the L layer (fresh litter). The leachable fraction of K at 120 h was 10 times higher than that of ^{137}Cs . Although additional studies are required to clarify the causes of the differences in the chemical forms of ^{137}Cs and K in litter, our result implies that the ^{137}Cs cycle in a forest, starting from the litter layer, is slower than that of K which provides a greater understanding of ^{137}Cs dynamics in forest litter layers. Consequently, it will provide important information on the management of contaminated forests by ^{137}Cs .

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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Appendix A. Supplementary data

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