



Simplified measurement method for dissolved radio-Cs in litter and soil seepage water using copper-substituted Prussian blue



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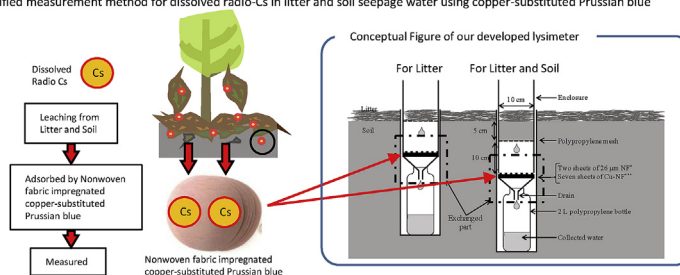
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HIGHLIGHTS

- We developed a simplified method for detecting dissolved radio-Cs in seepage water.
- 99% of dissolved ¹³⁷Cs from litter was collected in the field experiment.
- More than 90% of dissolved ¹³⁷Cs from soil seepage water was collected.
- This method reduces preparation time relative to conventional methods.
- The method can also separate the particulate and dissolved radio-Cs in-situ.

GRAPHICAL ABSTRACT

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ABSTRACT

We developed a simple and rapid method for detecting dissolved radio-Cs in litter and/or soil seepage water using nonwoven fabrics impregnated with copper-substituted Prussian blue (Cu-NF). In laboratory and field experiments, litter and/or soil seepage water including dissolved radio-Cs were passed through traditional lysimeter systems combined with seven sheets of the Cu-NF. We then examined the recovery ratios of dissolved ¹³⁷Cs in the Cu-NF. In the laboratory experiments with faster flow rates (11–2200 mm h⁻¹), over 86% of the total dissolved ¹³⁷Cs in litter seepage water was detected in the Cu-NF and over 82% of the collected ¹³⁷Cs was present in the first three sheets. In the field experiments, 99% of the total dissolved ¹³⁷Cs litter seepage water was collected in the Cu-NF and more than 96% of the collected ¹³⁷Cs was present in the first three sheets. Furthermore, the recovery ratio of dissolved ¹³⁷Cs increased with increasing installation Cu-NF length, probably because the packed soil in the Cu-NF lysimeter become more stable over time. Finally, because only the Cu-NF is measured, it is not necessary to undertake traditional measurement preparations such as filtration to remove particulate radio-Cs materials and evaporative concentration for low concentration of radio-Cs. As a result, we can save time and effort in measurement preparation by using the Cu-NF lysimeter method.

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1. Introduction

Large amounts of radionuclides were released into the environment during the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in 2011 (Endo et al., 2012) and were widely

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dispersed on the surrounding terrain. As forest areas account for more than 70% of land in the Fukushima Prefecture, radioactive cesium (radio-Cs), especially ^{137}Cs , will remain in the areas for a considerable period of time because ^{137}Cs has the long half-life of 30.17 years. Therefore, the radioactive contamination of forests and forest products will be a matter of serious concern for some time to come (Kuroda et al., 2013).

Soon after the FDNPP accident, radio-Cs deposited on the forest floors was estimated to migrate into soil by becoming dissolved in water; however, the transported amounts of dissolved radio-Cs rapidly decreased with time after the accident (Matsunaga et al., 2013). Dissolved radio-Cs can move rapidly from the litter layer into soil and then from the soil to plants (Nakanishi et al., 2013). Therefore, studies of the migration of the radio-Cs among these components (e.g. plants, litter layer and soil) are important over the coming decades, rather than focusing only on radio-Cs accumulation within each component (Hashimoto et al., 2012). In addition, clarification of radio-Cs dynamics in forest ecosystems will play an important role in applications such as forest management and radiation protection, taking account of the various uses of forest resources (Fujii et al., 2014).

Two different methods have been employed to evaluate vertical migration of radio-Cs in forest soil: the soil profile method and the lysimeter method. Many studies have investigated vertical migration of radio-Cs by using the soil profile method (e.g. Straume et al., 2006; Takahashi et al., 2015), in which migration rates were estimated from changes of radio-Cs in soil depth profiles during periods from 1 year to several years. Although this method is relatively simple, sampling point must be shifted slightly at each sampling period. As it was well known that radio-Cs in forest soil shows extensive spatial heterogeneity (Onda et al., 2015), the soil profile results may be affected by the spatial variation of radio-Cs in soil. There is also a possibility that evaluation over short periods is associated with substantial uncertainty.

In contrast, a lysimeter method directly monitors migrating ions in the soil (Rasmussen et al., 1986). Several types of lysimeter, such as a zero-tention lysimeter (Giesler et al., 1996) and suction lysimeter (Kosugi and Katsuyama, 2004) stainless steel lysimeter (Tegen and Dörr, 1996), had been developed for forest soil monitoring. Although these methods were used after the Chernobyl accident in 1986 (Tegen and Dörr, 1996) and the FDNPP accident in 2011 (Nakanishi et al., 2014), it has not been widely adopted for radio-Cs monitoring because of its complex procedure and a huge amount of effort for pretreatment and analysis of collected drainage water. For example, to employ the lysimeter method, we need to excavate soil deeply (tens of centimeters) to install a large sized lysimeter. Therefore, multi-point monitoring with lysimeters in a forest ecosystem is not practical. In addition, using the lysimeter method, we need to collect drainage water once in several weeks or a month and bring the water sample to a laboratory for radio-Cs measurement because the activity concentration of radio-Cs is typically low (Nakanishi et al., 2014). Determination of the radio-Cs concentration in the water sample requires filtration and evaporative concentration prior to measurement or a long period for a small volume of collected water or a very low concentration of radio-Cs. Furthermore, the conventional lysimeter method separates dissolved radio-Cs from particulate radio-Cs not in-situ but in the laboratory by filtration. Therefore, development of a simple, rapid and easy monitoring method for radio-Cs in litter and soil seepage water is required to concentrate dissolved radio-Cs on-site to be easily measured.

After the FDNPP accident, a rapid method was developed for the detection of dissolved radio-Cs in fresh water using nonwoven fabric impregnated with Prussian blue (Yasutaka et al., 2013) or potassium zinc ferrocyanide (Yasutaka et al., 2015). Prussian blue

(potassium ferrocyanide (II) potassium oxide iron (II), KFe $[\text{Fe}(\text{CN})_6]_3 \cdot x\text{H}_2\text{O}$) is known to specifically adsorb dissolved radio-Cs (IAEA, 1997). Yasutaka et al. (2016) also developed the nonwoven fabrics impregnated with copper-substituted Prussian blue (hereafter, Cu-NF) for detecting radio-Cs in seawater. The Cu-NF can absorb the radio-Cs in the water sample with high concentration of co-existing ions.

In this study, we aimed to develop a simple, rapid and in-situ separation method for examining the migration of dissolved radio-Cs from soil and/or litter in forest using Cu-NF. We introduced the Cu-NF to a conventional lysimeter, which can adsorb dissolved radio-Cs in the seepage water in-situ. We also examined the recovery ratio of dissolved radio-Cs with the Cu-NF method in both laboratory and field experiments.

2. Materials and methods

We conducted two experiments using Cu-NF combined with a lysimeter (hereafter termed the Cu-NF lysimeter): laboratory experiments and field experiments. First, we confirmed the absorption ability of dissolved radio-Cs in litter seepage water using the Cu-NF in laboratory experiments. Next, we conducted field experiments to examine the applicability of this method to litter and/or soil seepage water in the forest environment.

2.1. Experimental materials

The Cu-NF lysimeters for the laboratory and the field experiments consisted of PVC pipes (90 cm²), nonwoven fabric filters and a polypropylene bottle (I-Boy, As ONE Corporation, Japan), as shown in Fig. 1. Litter and/or soil were packed as packing materials in the PVC pipes, followed by two or three sheets of 26 μm pore size nonwoven fabric sheets (ELV-130 for the laboratory experiment, H-8010 for the field experiment, Japan Vilene Co., Ltd., Japan) and seven sheets of Cu-NF (volumetric density 0.03 g cm⁻³, thickness 0.01 cm, Japan Vilene Co., Ltd., Japan). The 26 μm pore size nonwoven fabric sheets were used to collect soil particles to avoid contamination of the Cu-NF. Both the Cu-NF and the 26 μm pore size nonwoven fabric were cut to a diameter of 113 mm. The water, passed through litter and/or soil, the 26 μm pore size nonwoven fabric sheets and the Cu-NF, was collected with 2 L polypropylene bottles.

2.2. Laboratory experiments

In the laboratory experiments, we evaluated the recovery ratio of dissolved radio-Cs in water passed through the litter in the Cu-NF lysimeter. The litter was separately collected from the L layer and F-H layer at a Japanese cedar forest (*Cryptomeria japonica*) in Kawamata, Yamakiya District, 40 km north-west of the FDNPP (37°36'29" N, 140°38'57" E; 583 m asl). Litter from the L layer (400 g) and the F-H layer (160 g) was mixed well after being cut to 2 cm in length. Next, the pipe was filled with litter (approximately 800 Bq/65 g wet weight) to a height of 5 cm in three of the Cu-NF lysimeters (Nos. 1-1, 1-2 and 1-3) and a control lysimeter (No. 1-4-1). In three of the Cu-NF lysimeters (Nos. 1-1, 1-2 and 1-3), two sheets of 26 μm pore size nonwoven fabric and seven sheets of Cu-NF were combined, and in the remaining lysimeter (No. 1-4-1), only three sheets of 26 μm pore size nonwoven fabric without Cu-NF was used as the control.

Table S1 and Fig. S1 show the experimental conditions and procedures. Ten liters of ion exchange water, which was approximately 1100 mm of throughfall volume and nearly the same as annual precipitation in Fukushima Prefecture (1166 mm, 1981–2010), was passed through each the Cu-NF lysimeter (Nos. 1-

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