



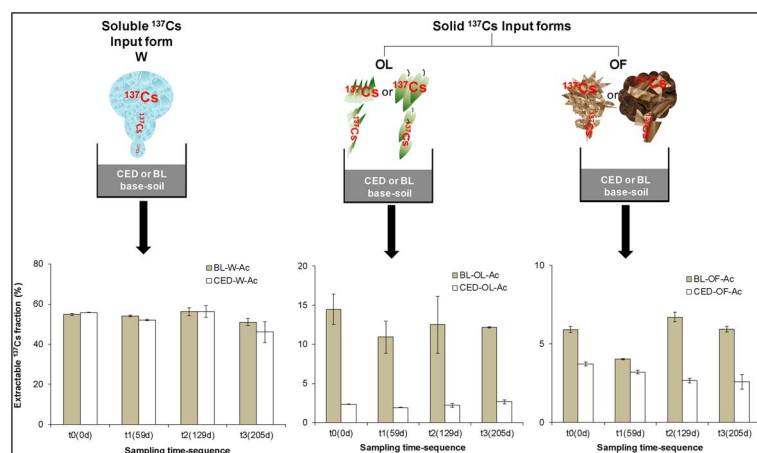
The impact of radiocesium input forms on its extractability in Fukushima forest soils

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GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:

Availability

¹³⁷Cs

Decomposition

Extractability

Litter

ABSTRACT

The effects of ¹³⁷Cs deposit forms on its ageing in soil have not yet been reported. Soluble and Solid ¹³⁷Cs input forms were mixed with the mineral soils collected under Fukushima's coniferous and broadleaf forests, incubated under controlled laboratory, and examined the evolution of ¹³⁷Cs availability over time. Results show that the extracted ¹³⁷Cs fraction with water was less than 1% for the soluble input form and below detection limit for the solid input forms. Likewise, with an acetate reagent, the extracted ¹³⁷Cs fraction ranged from 46 to 56% for the soluble input and from 2 to 15% for the solid input, implying that the nature of the ¹³⁷Cs contamination strongly influences its extractability and mobility in soil. Although the degradation of organic materials was apparent, its impact on the ¹³⁷Cs extractability was found to be weak. Nevertheless, more Ac-available ¹³⁷Cs was obtained from broadleaf organic material mixes than the coniferous counterparts, suggesting that the lignified nature of latter tend to retain more ¹³⁷Cs. When extrapolated to a field context, more available ¹³⁷Cs fraction may be expected from wet-derived contaminated forest soils than contaminated via solid-derived inputs. Such information could be helpful for radioecological management schemes in contaminated forest environments.

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1. Introduction

Following the Fukushima nuclear power plant accident, the forest-dominated large portion of the surrounding area was contaminated by the discharged radioactive materials. Among the released radionuclides, primarily due to its long half-life (30.1 y), ^{137}Cs became an important radioisotope deposit of interest, particularly in forest environments. Generally, the form of deposition that delivers radiocesium to soil can be categorized into two major carriers: *liquid carrier* and *solid carrier*. The liquid carrier group represents soluble ^{137}Cs input upon reaching the soil. This includes several liquids that range from precipitation forms to the solution of discharges of bulky ion mixtures. The second category, denoted as solid carrier, refers to radiocesium contaminated solid materials at the moment of deposition on soil. This form primarily receives radiocesium directly by intercepting ^{137}Cs deposits (dry/wet or both) or indirectly through secondary contamination pathways before it reaches the soil. Aerosol and falling contaminated plant organs, such as leaf litter, are in this category.

Several studies have been conducted at various scales (field-laboratory-modeling) to study the behavior of ^{137}Cs in soil, without specifically taking into account for its input forms [e.g., 1–3], and it has been concluded that the irreversible fixation of ^{137}Cs onto the soil particles is gradual and progressive over time. This process is commonly characterized by a decrease in the extractable fraction leading to a gradual increase in the fixed fraction, possibly born from specific exchange sites in clay soil particles - a process termed as *ageing* [4–6]. Concerning the role of clay soil particles on the distribution of radionuclides, a large volume of information is now available for the range of soil types, describing it as a radionuclide sink pool due to the presence of highly specific sorption site called Frayed Edge Sites (FES) [e.g., 7–15]. Unlike clay, the effects of soil organic matter (SOM) on ^{137}Cs ageing in the solid phase remain controversial between studies reporting a strong [e.g., 16–18] versus weak [19–22] fixation of radionuclides. Despite the argument, there is a general consensus that the upper organic soil section holds and retards the downward migration of radionuclides [23–29].

Moreover, studies highlighted that the depositional pattern of Fukushima-derived ^{137}Cs in a forest environment shifted from a throughfall dominated in the early period to a litterfall dominated transfer pathways in the latter period [3,30]. Hence, the contribution of soluble and litter input forms of ^{137}Cs to forest soils cannot be ignored, even in the periods after peak deposition. Moreover, as a part of the radiocesium transfer pathways, ^{137}Cs enriched litter layer (OL) and the fragmented layer (OF) in the forest floor remain imminent sources of ^{137}Cs contamination to all niches directly or indirectly connected to it, including the underlying mineral soil. Specifically, the downward migration of ^{137}Cs from this organic horizon to beneath the mineral soil is the resulting effect of very complex and interrelated biogeochemical processes operating in the soil such as organic matter degradation [31]. Hence, it is radioecologically an important soil section that has to be taken seriously. Bearing this in mind, no detailed studies on the effects of ^{137}Cs input forms on its extractability have been reported.

Consequently, unlike field observation, laboratory-based experiments can help to single out a particular process by limiting or eliminating other complex interactions and effect-overlaps, and can provide a platform to closely study the behavior of different forms of ^{137}Cs input into the mineral soil. Here, three forms of ^{137}Cs inputs (one soluble input, to represent a throughfall pathway, and two solid inputs-contaminated OL & OF layers, to represent litterfall and its subsequent eco-processed forms) were directly introduced into mineral soils obtained from two different forest types of the contaminated area around Fukushima. This study was aimed at evaluating the different ^{137}Cs input forms in soils and its subsequent availability (hereafter, the term is interchangeable with extractability) under the decomposition kinetics of organic matter.

Table 1

The physicochemical properties of the base soil used for the incubation experiment.

Experiment	Forest type		
	Broadleaf (BL)	Cedar (CED)	
Base soil			
Particle size (μm)	< 2	13.4 \pm 2.1	5.7 \pm 1.7
	2–20	37.5 \pm 4.6	24.5 \pm 3.0
	20–50	28.1 \pm 2.6	32.4 \pm 1.6
	50–200	14.4 \pm 5.9	30 \pm 4.0
	> 200	6.6 \pm 2.7	7.4 \pm 3.8
Main Mineral Components ^a			
	Quartz, anorthite, magnetite, kaolinite	Quartz, anorthite, magnetite, kaolinite	
C (mg g^{-1})	37.9 \pm 2.3	81.5 \pm 3.4	
C/N	7.3 \pm 1.1	8.7 \pm 0.8	
pH _{H₂O} (1:5) (s/w)	4.7 \pm 0.1	5.3 \pm 0.1	
Added organic material			
OL	C (mg g^{-1})	420 \pm 41	437 \pm 44
	C/N	33 \pm 2	41 \pm 4
	pH _{H₂O}	4.9 \pm 0.2	6.0 \pm 0.2
OF	C (mg g^{-1})	392 \pm 22	347 \pm 35
	C/N	20 \pm 2	21 \pm 2
	pH _{H₂O}	4.7 \pm 0.3	6.1 \pm 0.3

^a Coppin et al. [32].

2. Material and methods

2.1. Soil sampling

Representative sampling plots were selected under broadleaf (hereafter BL) and coniferous forests of Japanese cedar (hereafter CED) located at about 40 km northwest from the Fukushima nuclear power plant. The OL and OF layers were carefully separated by hand. Their respective underlying mineral soils were also carefully collected layer by layer at depth intervals of 0–3 cm, 3–8 cm and 8–20 cm. Detailed information is given by Coppin et al. [32].

2.2. Properties of base soil and organic materials

The base soil here refers to the uncontaminated mineral soils (8–20 cm) to which different ^{137}Cs input forms were applied. The organic materials refer to the OL and OF, which were used as solid ^{137}Cs input forms (for detailed information see Table 1 and Supplementary text: Material and methods).

2.3. Preparation of a sample mixture and ^{137}Cs contamination

2.3.1. Soluble ^{137}Cs input form

Based on the available soil materials, 340g and 335g (dry mass) of mineral soil from BL and CED, respectively, were prepared. The spiking solution was made by diluting a total of ~ 4 MBq (9.2×10^{-9} M) ^{137}Cs in pure water in the presence of 2×10^{-3} M NaOH, to reach a pH of 5.40. A thin slice of soil was placed in a beaker, saturated with ultra-high quality water (UHQ) and contaminated uniformly by the spiking solution dripped carefully onto the whole surface, and then covered by a new thin soil slice and contaminated in the same way. This protocol was repeated until all of the soil and spiking solution were used up (~ 2 MBq equivalent to 4.6×10^{-9} M per soil type).

Based on their respective field moisture contents defined at the time of sampling, the contaminated-water-saturated soils were air-dried until the final moisture contents reached about 41% (BL) and 60% (CED) soils, and then well-mixed to homogenize them. This gave an initial contamination rate of about 6.0 ± 0.2 kBq g^{-1} for each sample set. After taking the samples for time zero, the mixtures were transferred into a 0.5 L O-ring rubber septum screw cap jar, in duplicate per forest type, and then connected to the incubation chamber (Fig. 1).

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