Extractability of 137Cs in Response to its Input Forms into Fukushima Forest Soils

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Abstract

In case of nuclear accidents like Fukushima disaster, the influence of 137Cs depositional forms (soluble and/or solid forms) on mineral soil of forest environment on its availability have not reported yet. Soluble (137Cs tagged ultra-pure water) and solid (137Cs contaminated litter-OL and fragmented litter-OF) input forms were mixed with the mineral soils collected under Fukushima coniferous and broadleaf forests. The mixtures then incubated under controlled laboratory condition to evaluate the extractability of 137Cs in soil over time in the presence of decomposition process through two extracting reagents- water and ammonium acetate. Results show that extracted 137Cs fraction with water was less than 1% for soluble input form and below detection limit for solid input form. On the same way with acetate reagent, the extracted 137Cs fraction ranged from 46 to 56% for soluble input and 2 to 15% for solid input, implying the nature of 137Cs contamination strongly influences the extractability and hence the mobility of 137Cs in soil. Although the degradation rate of the organic materials has been calculated in the range of 0.18 ± 0.1 to 0.24 ± 0.1 y-1, its impact on 137Cs extractability appeared very weak at least within the observation period, probably due to shorter observation period. Concerning the treatments of solid 137Cs input forms through acetate extraction, relatively more 137Cs has been extracted from broadleaf organic materials mixes (BL-OL & BL-OF) than the coniferous counterparts. This probably is due to the fact that the lignified coniferous organic materials (CED-OL & CED-OF) components tend to retain more 137Cs than that of the broadleaf. Generally, by extrapolating these observations in to a field context, one can expect more available 137Cs fraction in forest soil from wet depositional pathways such as throughfall and stemflow than those attached with organic materials like litter (OL) and its eco-processed forms (OF).

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1. Introduction

In fact following the bomb test, Chernobyl and Fukushima nuclear accidents, several literatures are become available, which have reported the behavior and various aspects of different anthropogenic radionuclides including radiocesium in various ecosystem media based on either from field monitoring data or/and modelling approaches (e.g., Bunzl et al., 1989, 2000; Calmon et al., 2015; Coppin et al., 2016; Fujiyoshi and Sawmura, 2004; Gonze and Calmon, 2017; Kato et al 2017; Onda et al., 2015; Ruhm et al., 1996; Schimmack and Schulz, 2006). Moreover, a large volume of information is now available for the range of soil types, describing the role of clay soil particles on the distribution of radionuclides as a main sink pool (e.g., Campbell and Davies, 1995; Cremers et al., 1988; Hou et al., 2003; Hsu and Chang, 1994; Lehto, 2015; Saito et al., 2014; Valcke and Cremers, 1994). **However, the influence of ^{137}Cs depositional forms (soluble and/or solid forms) on forest mineral soil against its availability or ageing have not yet reported.**

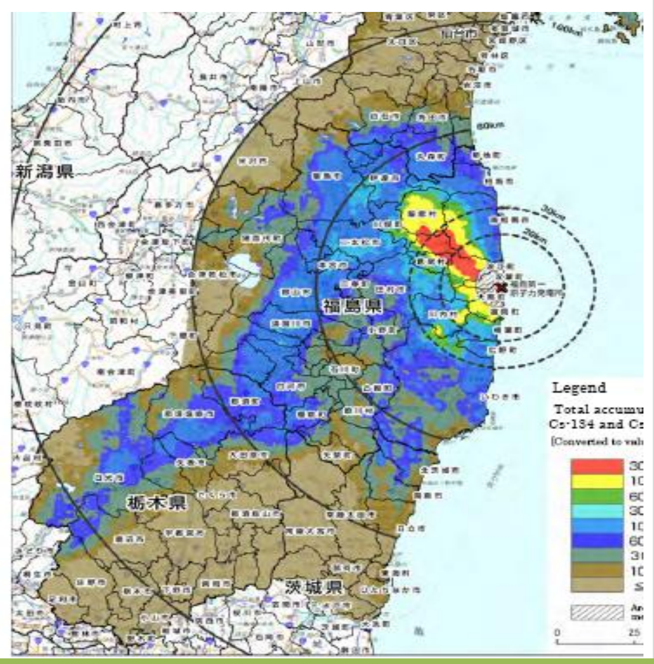
The forms of ^{137}Cs deposition on soil can be generally categorized in two as Liquid and Solid carriers. The liquid carrier group includes several liquids coming from precipitation forms (e.g., rainfall, throughfall, stemflow). The second category, denoted as solid carrier, refers to radiocesium contaminated 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 (e.g., translocation, root-uptake, re-suspension) before it reaches the soil. Aerosol and falling contaminated plant organs, such as leaf and branch litter, are in this category.
As at field condition, it is hard to separate ^{137}Cs input forms due to complex interaction and effect-overlaps, laboratory experiment can help us to single out particular factor and provide an immerse chances to closely examine its role and effects. Therefore, a laboratory based study was conducted to examine the impact of ^{137}Cs input form onto Fukushima forest soils on its availability.

2. Objective

- ❑ To evaluate the extractability of ^{137}Cs from forest mineral soils contaminated by solid (litter, OF) and liquid (water) forms of ^{137}Cs inputs under the presence of decomposition process.

3. Material and Methods

3.1. Sample collection



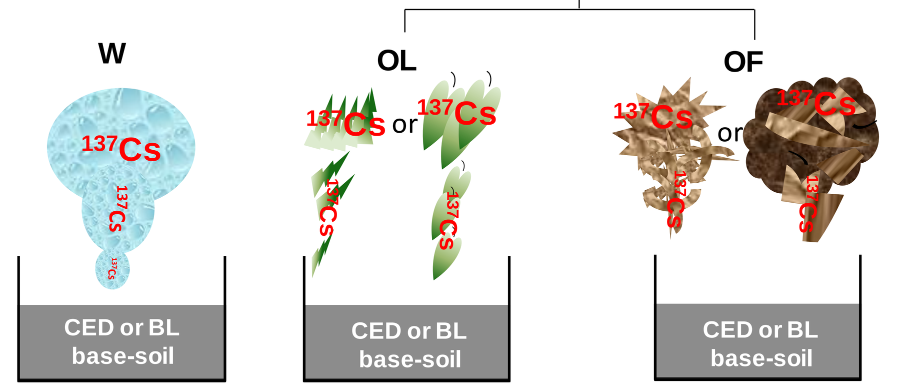
Map of radiation plume and contamination degree around the crippled Fukushima nuclear power plant around the study site

Both uncontaminated mineral soils, and contaminated OL and OF materials were carefully separated and collected from representative plots under Broadleaved (*Quercus serrata*) and Japanese Cedar (*Cryptomeria japonica*) forest stands, located in the contaminated forest areas of Fukushima prefecture, North-Eastern Japan. In this study, the Broadleaved and Japanese cedar are abbreviated as BL and CED, respectively. Moreover, the uncontaminated mineral soil collected from each forest stand represents their respective base mineral forest soils.

The detail description of the sites and sampling procedures are given by Coppin et al. (2016) and Kato et al., (2017).

3.2. Laboratory procedures

Mixing with contaminant & homogenize → Incubating → Periodically sampling from incubated bottles



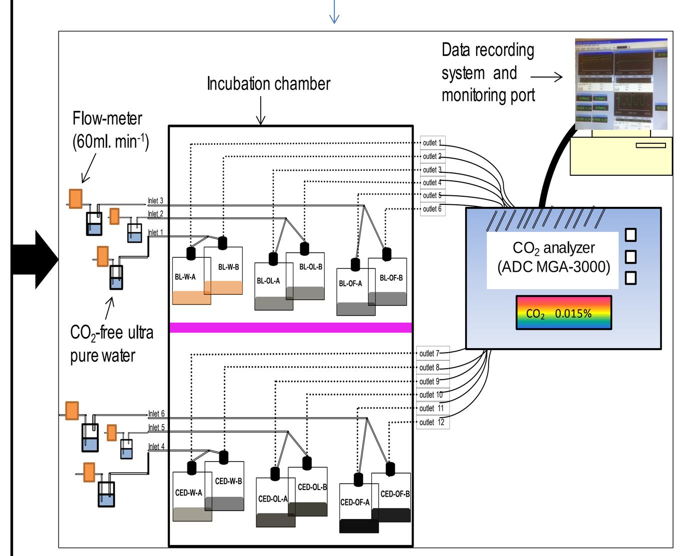
Base soil to OL,OF mix ratios
BL: 7:1 for both OL&OF
CED: 2:1(OL), 1.3:1(OF)

Base soil property

	BL	CED
%Clay	13	6
%C	3.8	8.2
C/N	7.3	8.7
pH	4.7	5.3

$^{137}Cs_0$ contamination rate of the mixtures
BL&CED-W: 6 kBq/g
BL: 2.4(OL), 13(OF) Bq/g
CED: 12 (OL), 35(OF) Bq/g

Some properties of the mixtures
%C: BL-W = 3.8, BL-OL = 9.5, BL-OF = 9
CED-W = 8.2, CED-OL &OF = 22
C/N: BL-W =7, BL-OL = 14, BL-OF = 13
CED-W = 9, CED-OL = 22, CED-OF = 16
pH: BL-W = 5.3, BL-OL&OF = 5.7
CED-W = 5.8, CED-OL = 5.8, CED-OF = 6.3



10g $_{\text{dry mass eqv.}}$ for
- %MC
- Bq/g
- %C &
- C/N measurements

3-6g $_{\text{dry mass eqv.}}$
For 4-step successive ^{137}Cs extractions

↓

Mixed with 30 – 120ml (water or Acetate)

↓

Shaken for 2-24hrs (300 rmp)
Centrifuged for 25min (10,000xg)

↓

Extracted with 0.8µm filter (repeated 4x)

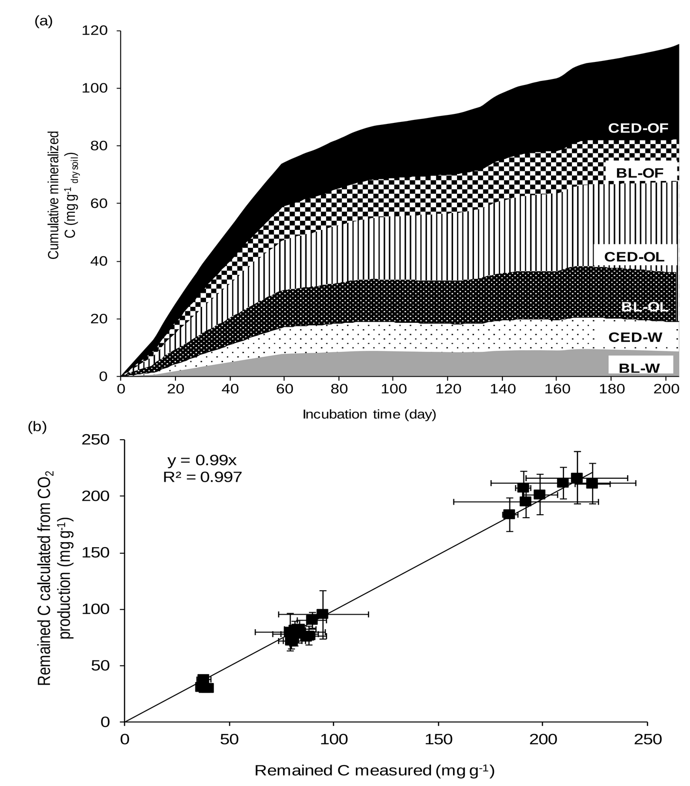
→ **^{137}Cs measured by γ-spectrometer**

3.3. Estimation of the available ^{137}Cs fraction

The results of % of available ^{137}Cs fraction was obtained by four step extraction procedure.

4. Results and Discussion

4.1. Carbon mineralization



- Carbon (C) mineralization in the system varied among the treatment units due to
  - The initial decomposable (labile C) materials differences (Artz et al., 2006).
- The order of C mineralization rate was :

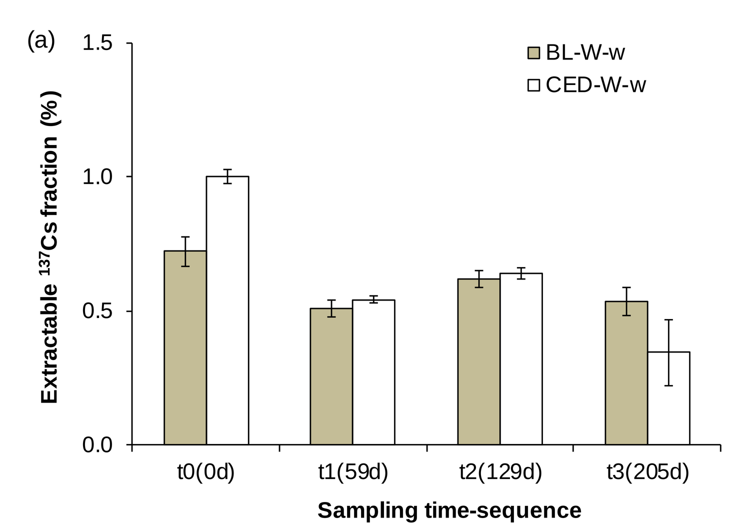
CED-OF,CED-OL > BL-OL > BL-OF > CED-W > BL-W

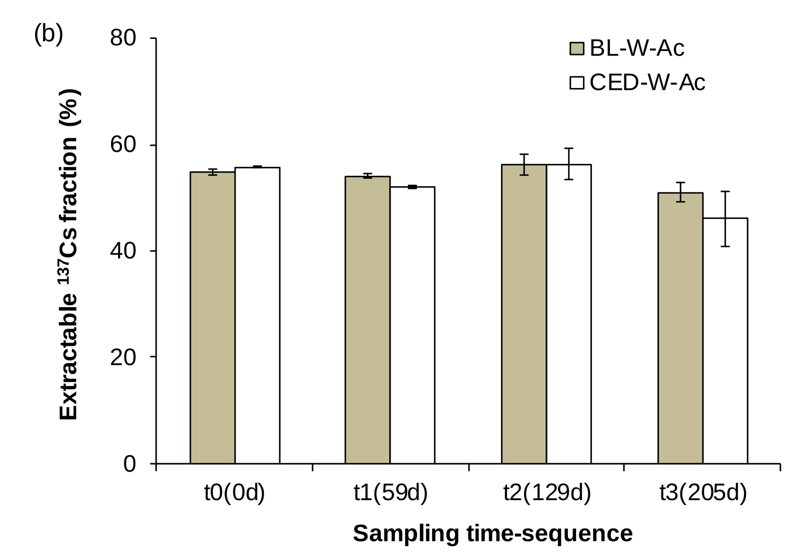
The degradation amount and rate of the added organic materials (OL and OF) were:
- ➢ BL-OL = 14% (0.24 y^{-1})
- ➢ BL-OF = 10% (0.18 y^{-1})
- ➢ CED-OL = 13% (0.23 y^{-1})
- ➢ CED-OF = 13% (0.23 y^{-1})

Our results agree with previous studies obtained from litterbag experiments in the field (Shutou and Nakane, 2004; Salamanca et al., 1998).

4.1. Extractable ^{137}Cs fraction

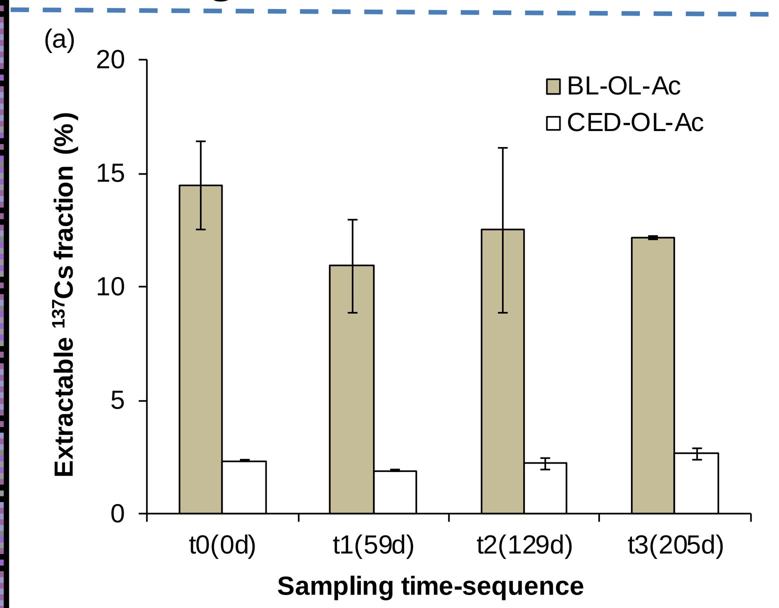
Liquid input

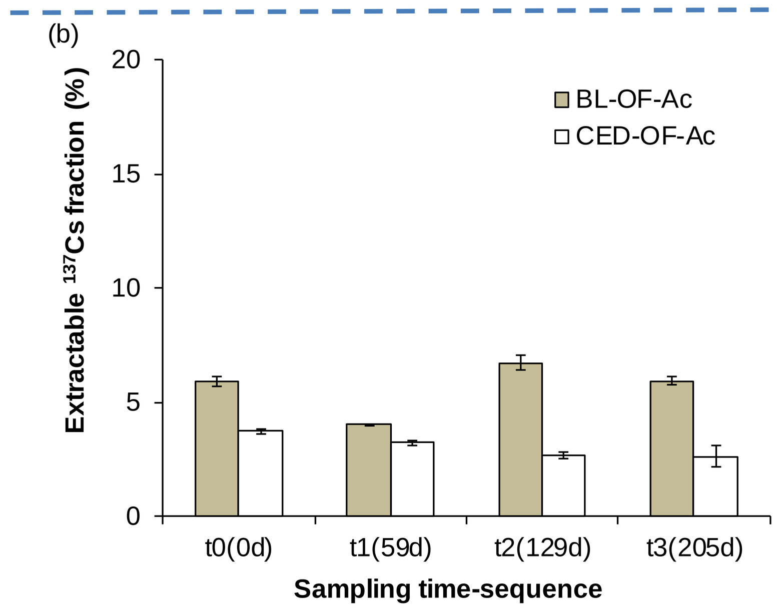




- w-available ^{137}Cs fractions (Fig. a) were found to be low for both forest types across the incubation period. Bouzidi et al. (2010), Saito et al. (2014) and Takeda et al. (2013), collectively indicated that only small amounts of ^{137}Cs are water soluble, which are probably generated from weakly binding sites.
- Ac-available (Fig. b) , > 50% of the ^{137}Cs fraction was consistently made available from both forest soils, probably due to additional processes, generated by the chemical nature of the reagent.
- Only small difference was observed along the incubation time course.

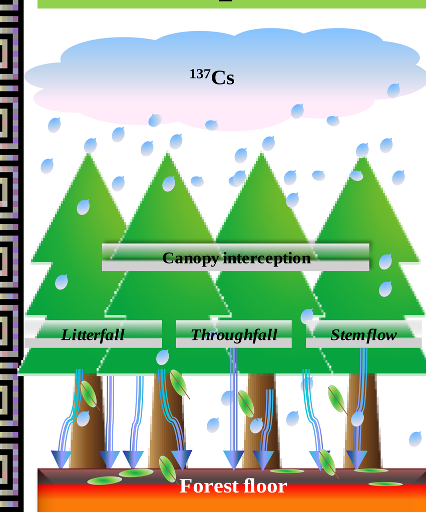
Solid input





- w-available ^{137}Cs fractions from the OL and OF input treatments were close to the detection limit with high uncertainties.
- Ac-available ^{137}Cs fraction (3-15%) is lower than the liquid input forms (>50%).
- More Ac-available ^{137}Cs fraction was generally: BL > CED and from OL > OF (Fig. a, b), probably due to higher lignin content of the coniferous litter that retain more ^{137}Cs than broadleaf deciduous litters (Hashida and Yoshihara, 2016; Nakanishi et al., 2014).

4.3. Implications



- Despite small and last short of w-available ^{137}Cs fraction, it is responsible in defining the initial ^{137}Cs depth profile in the field (Koarashi et al., 2012; Teramage et al., 2014, 2016).
- Taking the Ac- available fraction, > 50% ^{137}Cs could be bioavailable in the field during wet-derived deposition.
- As litterfall remains the as a dominant process in latter period, ^{137}Cs is more likely to stay in the biogeochemical active system of the forest floor (Kato et al., 2017; Yoschenko et al, 2017)

5. Conclusions

- ❑ ^{137}Cs input forms influenced its extractability in soil and the w-extractable fraction was very low despite the input forms.
- ❑ Ac- extractable ^{137}Cs fraction was high for liquid input form compare to solid, and for the latter it is higher in BL than CED.

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