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**Impact of forest thinning on the dynamics of litterfall derived <sup>137</sup>Cs deposits in coniferous forest floor after Fukushima accident**

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**Résumé d'auteur** : The effects of a 50% forest thinning intensity on Fukushima-derived <sup>137</sup>Cs deposition by litterfall and its discharge by runoff in hillslope coniferous forest were monitored using four litterfall traps and a hillslope erosion plot. The observation was underway during the pre-and post-thinning periods. Results demonstrated that during the pre-thinning period a total  $150 \pm 13 \text{ g m}^{-2}$  of litterfall deposited about  $924 \pm 69 \text{ Bq m}^{-2}$  of <sup>137</sup>Cs. This accounts for 11% of the local <sup>137</sup>Cs fallout recorded for the study site in the aftermath of the accident. After thinning, both litterfall and <sup>137</sup>Cs increased by more than six- and twofold, respectively. This is possibly owing to the slow individual tree recovery rate assisted by the change on the running space provided by canopy openings, which can accelerate even the normal gust wind to gain damaging power on the unshielded mechanically injured parts of the contaminated residual trees. In both cases, litterfall generally transferred about 37% ( $3 \pm 0.2 \text{ kBq m}^{-2}$ ) of the local <sup>137</sup>Cs fallout onto the forest floor over the observation period. The eroded litter-associated <sup>137</sup>Cs increased by about a factor of two after thinning, which only accounted for less than 1% of <sup>137</sup>Cs deposited by litterfall. This implies that the forest floor retains <sup>137</sup>Cs and remains contaminated regardless of the size of the eroded litter material. But this could become a potential secondary contamination source for the downstream resources such as water bodies and villages, especially at the time of flooding, which in turn calls a serious attention in designing decontamination schemes.

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## 1. Introduction

Several studies have indicated that soil water content affects the availability of radionuclides (e.g., Degryse et al., 2004; Sohlenius et al., 2013; Tanaka and Yamamoto, 1991). Rosen et al. (2006) showed that succession of dry and wet cycle compared to wet conditions increased the bioavailability of  $^{134}\text{Cs}$  that generated a greater uptake by rye grass from organic soil. Popov et al. (2014) also demonstrated that the addition of 5% organomineral sorbent (OMS) on sandy soddy-podzolic soil increased the effect of the drying and wetting cycles on the selective sorption of radiocesium ( $^{137}\text{Cs}$ ). Therefore, depending on the binding strength,  $^{137}\text{Cs}$  in soil can be available and mobile by rain, soil water as well as exudates secreted by plants. In a laboratory incubation experiment using constant soil moisture content, Teramage et al. (2018) recently demonstrated that the extractability of  $^{137}\text{Cs}$  in forest soils depends on its input forms (i.e., aqueous  $^{137}\text{Cs}$  vs. solid organic sources of  $^{137}\text{Cs}$ ). They found that both the water- and acetate-extractable  $^{137}\text{Cs}$  fractions were greater for aqueous amended soils than for solid organic amended soils. Results show that the extracted  $^{137}\text{Cs}$  fraction with water was less than 1% for the aqueous amended soils and below detection limit for the solid organic amended soils. Likewise, with an acetate reagent, the extractable  $^{137}\text{Cs}$  fraction ranged from 46 to 56% for aqueous amended soils and from 2 to 15% for the solid organic amended soils.

Indeed, in field environments, the soil moisture content fluctuates depending on the seasons, climatic conditions and unforeseen weather situations such as tropical cyclones, which can modify the solution/solid elemental distribution. In Japan, including in the contaminated Fukushima region, the fluctuation of soil moisture triggered by seasonal changes, floods and typhoons followed by dry weather events, is a commonly occurring phenomenon. Therefore, studies on the

impact of drying and wetting cycles in the laboratory can provide information on the behavior of  $^{137}\text{Cs}$  in soils of varying moisture contents. The objective of this study was to complete the previous study (i.e., Teramage et al., 2018) by including and investigating the impact of drying and wetting cycles on the  $^{137}\text{Cs}$  availability in soils.

## 2. Material and methods

The full description of the incubation experiment, including the incubation set up, experimental units and conditions, sampling schemes and sample analysis has been described elsewhere (Teramage et al., 2018). In short, the contaminated litter (OL) and fragmented (OF) layers were carefully separated by hand while the underlying soils at a depth of 8 – 20 cm representing uncontaminated mineral soils, were collected under both broadleaf (BL) and cedar (CED) forests in the contaminated Fukushima area in Japan. The type of both soils used was andosol with a silty-loam texture. Although micaceous minerals were not detected, the clay content was about  $13\pm 2\%$  in BL and  $6\pm 2\%$  in CED soils composed of quartz, anorthite, magnetite and kaolinite (Coppin et al., 2016). The carbon content of the soil in the CED forest ( $8.2\pm 0.3\%$ ) was more than twice that in the BL ( $3.8\pm 0.2\%$ ). The pH of both soils was acidic (CED:  $5.3\pm 0.04$ , BL:  $4.7\pm 0.01$ ). The carbon content in the added organic materials ranged from  $35\pm 4\%$  (CED-OF) to  $44\pm 4\%$  (CED-OL).

The respective mineral soils were contaminated by mixing either  $^{137}\text{Cs}$ -contaminated water, BL-W and CED-W (initial contamination rate of  $\sim 6.0 \pm 0.2 \text{ kBq g}^{-1}$  for both BL and CED, carrier  $^{133}\text{Cs}$  addition represent less than 1% of  $^{133}\text{Cs}$  content in soil) or  $^{137}\text{Cs}$  contaminated OL/OF materials. Based on the availability of the OL and OF materials and to keep the carbon content as close as to its content in the top soil layers at the field (Coppin et al., 2016), a ratio of about 7:1 (dry mass) mineral soils to  $^{137}\text{Cs}$  contaminated OL or OF for BL and 2:1 (OL) and 1.3:1 (OF) for CED were thoroughly mixed (initial contamination rates of  $2.4\pm 0.2 \text{ Bq g}^{-1}$  (BL-OL),  $13.0\pm 0.7 \text{ Bq}$

$\text{g}^{-1}$  (BL-OF),  $12 \pm 0.04 \text{ Bq g}^{-1}$  (CED-OL) and  $35 \pm 0.03 \text{ Bq g}^{-1}$  (CED-OF)). The six respective contaminated mixtures (BL-W, -OL, -OF and CED-W, -OL, -OF) were transferred into incubation bottles (130 to 200 g dry weight) with two replications and the experiment set in motion.

The moisture regime of the incubated samples over the entire observation period is illustrated in Figure 1. As indicated in Figure 1, after completing the constant moisture incubation experiment ( $\sim 40\%$  for BL and  $\sim 60 - 80\%$  for CED, defined by the field moisture contents at the time of sampling), the remaining samples were exposed to four complete drying-and-wetting cycles (i.e. approximately 40-day cycles) starting from 286 days after incubation. The moisture content fluctuated from very wet (BL: 134% and CED: 230%) to nearly dry (both conditions:  $\sim 2\%$ ). Samples from the incubated bottles were taken to monitor different parameters after three complete dry and wetting cycles from all experimental units (i.e. after 377 days of incubation) and after the fourth complete cycle (i.e. after 440 days of incubation) only for the liquid contamination treatment for BL conditions (due to insufficient remaining samples in the BL-OL and BL-OF treatments). At each sampling time, samples of about 3 g (from aqueous contamination) and 6 g dry mass (from solid contamination) for successive extraction and about 10 g dry mass (from each contamination type) were taken to monitor the  $^{137}\text{Cs}$  activity, moisture and carbon contents.

The successive extraction technique was carried out in tubes containing desorption reagents to soil ratio of  $10\text{ml g}^{-1}$  for BL-W and CED-W and  $20\text{ml g}^{-1}$  for BL-OL, BL-OF, CED-OL and CED-OF treatments. The tubes were then shaken for 24h (water) or 4h (acetate reagent) and centrifuged to separate the supernatants. Thus, the same process was repeated four times, refilling the residue with a similar amount of each reagent, and fully dispersing the soil at the beginning of each stage. The extracted  $^{137}\text{Cs}$  concentrations at each four successive extractions were used to estimate the coefficient of distribution,  $k'_d$  (as the slope of the straight line) of the extractable fraction and the

fixed  $^{137}\text{Cs}$  fraction in solid materials (as the y-intercept) from the graph plotting the final concentration of  $^{137}\text{Cs}$  in the solids against the final  $^{137}\text{Cs}$  concentration in the solution obtained at each extraction stage. The detailed sampling and the analysis of the various parameters were the same as described by Teramage et al. (2018). ANOVA at the 0.05 significance level was used for statistical analysis and comparison whenever required.

### **3. Results and discussion**

#### **3.1. Dynamics of organic carbon degradation**

The evolution of organic carbon content ( $C_{\text{org}}$ ) in soils contaminated with water or organic amendments (with OL or OF) are provided in Figure 2. A single first order kinetic law can explain the  $C_{\text{org}}$  degradation over time, suggesting the same degradation rate throughout our experiments. Compared to the soils contaminated from liquid input, for which the  $C_{\text{org}}$  degradation was very weak, the  $C_{\text{org}}$  degradation rate was higher in the soils contaminated by the organic materials particularly for the OL conditions. This is obviously due to the increase of the degradable plant material (DPM), which constitutes a labile carbon pool by the added organic materials (Jenkinson et al., 1992). For each treatment, the magnitude of  $C_{\text{org}}$  degradation kinetics remained unaffected by the drying and wetting cycles at least during the observation period. Indeed, several studies have indicated that an optimum moisture, which is usually somewhere near field capacity (i.e. macropore space air-filled and micropore spaces water-filled), is required for the decomposer communities in the soil to promote during  $C_{\text{org}}$  degradation (e.g., Chen et al., 2000; Davidson et al., 2000; Gabriel and Kellman, 2011; Ilstedt et al., 2000; Moorhead et al., 1999). Linn and Doran (1984) have also indicated that the microbial respiration decreased both in very wet and dry soils due to limited  $\text{O}_2$  diffusion in soil pore spaces and reduction soluble organic carbon substrates in water films, respectively. However, Cortufo et al. (2010) have reported that the degradation rate

of *Arbutus unedo* leaf litter was higher for wet treatments (Mean residence time-MRT: 2.99 years) and lower for dry treatments (MRT: 4.26 years) compared to the control treatment (MRT: 3.23 years) in a laboratory incubated experiment. Borken and Matzner (2008) also explained that the additional mineralization during the wetting of soil could be associated with several processes such as exposure of previously protected organic matter by wetting pulse, while frequent oscillating soil moisture diminishes the wetting pulse and hence the degradation. However, this was not the case in our observation, where the trends of  $C_{org}$  degradation were not affected by drying and wetting cycles. This is possibly either due to the presence of diverse microbe communities which become active at both extreme moisture conditions that compensated and maintained the  $C_{org}$  degradation, or our observation time was not sufficient for the effect to manifest. Another explanation could be that the choice of the initial soil moisture content close to the field capacity resulted in optimum  $C_{org}$  degradation, and the oscillated degradation rate due to the drying and wetting cycle averaged out to keep this optimum value.

### **3.2. Impact of drying and wetting on the availability of $^{137}Cs$ and $k'_d$**

The evolution of  $^{137}Cs$  in terms of water-extractable fraction (w-available) for the liquid contamination only, the acetate-extractable fraction (Ac-available) for both contaminations and the associated distribution coefficient ( $k'_d$ ) during the entire observation period, including the drying and wetting treatment is given in Table 1. Except for the results related to drying and wetting treatment, the rest of the observations have been discussed in detail elsewhere (Teramage et al., 2018). The information is provided here for further comparison and discussion of the results related to the impact of drying and wetting cycle treatments.

#### ***3.2.1. Soils contaminated by liquid $^{137}Cs$ input***

For this contamination treatment, the  $k'_d$  values for both water- and acetate-extractable  $^{137}\text{Cs}$  fractions in the BL and CED soils remained nearly the same with time except for a relatively high value at the initial stage for the water-extractable fractions (Table 1). This was possibly due to pseudo or temporary retention of  $^{137}\text{Cs}$  on to the soil particles at zero contact time. The  $k'_d$  values were also unaffected by the drying and wetting cycle treatments, indicating the dynamics of  $^{137}\text{Cs}$  distribution between the solution and the solid phase is similar regardless of the time and treatments. The water-extractable  $^{137}\text{Cs}$  fraction for both BL and CED was very small with a general decreasing trend, including values obtained after the drying and wetting cycle treatments (Table 1). This fraction decreased from  $1.00\pm 0.03\%$  to  $0.29\pm 0.03\%$  for CED and from  $0.72\pm 0.03\%$  to  $0.29\pm 0.01\%$  for BL at the beginning and at the end of the observation period, respectively. By taking the whole observation data as illustrated in Figure 3, the rate constant of the water-extractable  $^{137}\text{Cs}$  fraction was calculated as  $4.4 \times 10^{-3} \text{ d}^{-1}$  for CED and  $1.7 \times 10^{-3} \text{ d}^{-1}$  for BL.

As expected, the acetate-extractable  $^{137}\text{Cs}$  fraction was higher than the water-extractable fraction and decreased with time. The trend was similar for the two different periods, constant moisture or drying and wetting cycles, with a decrease from 55% to 30% for BL and 56% to 37% for CED. This implies that drying and wetting cycles weakly affects the  $^{137}\text{Cs}$  availability and its progressive ageing in time. In good agreement, Rosen et al. (2006) reported that the acetate-extractable  $^{137}\text{Cs}$  fraction across eight drying and wetting cycle treatments applied to contaminated peat and peaty podzol soils in the laboratory experiment was nearly the same across the cycles, confirming no clear effect of the drying and wetting cycles. Though not a direct desorption study, Tanaka and Yamamoto (1991) shown that drying had no influence on  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  concentrations in the top layers of sandy soil in a laboratory experiment. Taking the data, including the drying and wetting treatments, the calculated rate constant values for acetate-extractable fraction were relatively lower

(BL:  $8.2 \times 10^{-4}$ ; CED:  $9.4 \times 10^{-4} \text{ d}^{-1}$ ) than that of their water-extractable counterparts (Figure 3). Statistically, both the water-and acetate-extractable  $^{137}\text{Cs}$  fractions were not significantly different between BL and CED for their respective treatment types (Single factor ANOVA,  $P = 0.918936$  for water-extractable BL vs. CED;  $P = 0.852618$  for acetate-extractable of BL vs. CED), which indicates that water-derived  $^{137}\text{Cs}$  contamination pattern is not influenced by the small differences existing between these soils, mainly related to the OM content. Furthermore, the general decreasing pattern observed for the water-and acetate-extractable fraction in both forest soils indicates that the stabilization of water-and acetate-extractable fractions are not responsive to the three and four drying and wetting cycle treatments.

### **3.2.2. Soils contaminated by solid $^{137}\text{Cs}$ inputs**

Saki et al. (2015) obtained a measurable leached  $^{137}\text{Cs}$  amount from contaminated litter soaked alone in water than that soaked in water with minerals. However, in our observation it is clear that the water-extractable  $^{137}\text{Cs}$  fraction for this contamination treatment was below a measurable amount, even after drying and wetting cycles. Both low  $^{137}\text{Cs}$  in the organic contaminated material or desorption and resorption of  $^{137}\text{Cs}$  by mineral particles in the way beyond the water desorption capacity is highly probable, which would result in no water-extractable  $^{137}\text{Cs}$  (Teramage et al., 2018), and hence it is not discussed here.

The  $k'_d$  values of the acetate-extractable  $^{137}\text{Cs}$  fraction remained nearly the same across the observation time, except for low values observed at time zero (Table 1). The lower  $k'_d$  values at the initial observations may indicate the tendency of more acetate-extractable  $^{137}\text{Cs}$  fraction present in the solution phase from less tightly bound fractions on the surface of the organic materials. According to the observation, there were no definite effects of drying and wetting cycle treatments on the  $k'_d$  values.

Compared to the acetate-extractable  $^{137}\text{Cs}$  fraction from the soils contaminated with liquid  $^{137}\text{Cs}$  input, the acetate-extractable  $^{137}\text{Cs}$  fraction from the soils contaminated with solid (OL and OF) $^{137}\text{Cs}$  inputs was generally very low and practically unchanged with time, implying the nature of contamination affects the  $^{137}\text{Cs}$  availability and the absence of further  $^{137}\text{Cs}$  ageing. Further, the stability of the acetate-extractable  $^{137}\text{Cs}$  fraction from the soils contaminated with solid is in line with the absence of impact of drying and wetting treatment on the carbon degradation as the same observed in the constant moisture treatments. The acetate-extractable  $^{137}\text{Cs}$  fraction was larger in BL-OL (11 – 17%) and BL-OF (4 – 7%) than in their CED counterparts (2 – 5%). Statistically, the acetate-extractable  $^{137}\text{Cs}$  fraction in BL-OL treatment was significantly different from the BL-OF ( $P = 0.00017$ ), CED-OL ( $P=0.0000039$ ) and CED-OF ( $P=0.0000036$ ) treatments, highlighting that the acetate-extractable  $^{137}\text{Cs}$  fraction from the contaminated litter tends to depend on the litter types and its degradation status. In connection to this, several studies from laboratory and field conditions have revealed that the leaching and migration of  $^{137}\text{Cs}$  in deciduous forest floors are greater than from coniferous floors (e.g., Hashida and Yoshihara, 2016; Koarashi et al., 2016; Kurihara et al., 2018; Nakanishi et al., 2014). For example, Koarashi et al. (2016) reported that approximately 80% of the deposited  $^{137}\text{Cs}$  in deciduous broad-leaved forest migrated to minerals located below the organic layer, whereas more than half of the deposited  $^{137}\text{Cs}$  remained in the organic layer of the coniferous forest. Kurihara et al (2018) also observed faster  $^{137}\text{Cs}$  output flux from decomposing broadleaf forest litter than from the cedar forest litter. These collectively confirm that the litter type does affect the  $^{137}\text{Cs}$  dynamics in soil.

Rigol et al. (1999) reported a 2-3-fold decrease of exchangeable  $^{137}\text{Cs}$  fraction after drying and wetting cycles for 9 months in podsoles and peaty-podsoles. Moreover, Park et al. (2018) have reported that repeated drying and wetting cycles changed expandable clay minerals (e.g.

vermiculite) to non-expandable structure (e.g. illite like minerals) which drastically reduced the available Cs fractions. However, in our observation (where the clay minerals dominated by kaolinite), the impact of drying and wetting cycles had statistically no effect on the acetate-extractable  $^{137}\text{Cs}$  fraction, which can be associated with the nature of the clay minerals. In fact, an increase of approximately 28% of the acetate-extractable fraction in the BL-OL condition was observed after three complete cycles (17% at t-377) as compared to the immediate value before the treatment (12.2% at t-205) (Table 1). However, as the shape of the curve didn't change from the trend of the regular moisture treatment, it was hard to confirm that the observed result was due to the effect of the drying and wetting treatment, and not to the incubation time.

#### **4. Conclusion**

If taking the entire observation period into account, including the incubation at constant moisture and the incubation during drying and wetting cycles,  $^{137}\text{Cs}$  ageing was observed in soils contaminated with liquid  $^{137}\text{Cs}$  input whereas this was not the case for soils contaminated with solid  $^{137}\text{Cs}$  inputs. Nevertheless, it is possible to conclude that the  $^{137}\text{Cs}$  availability in soil likely depends on the nature of contamination, the litter type and a certain extent on its degradation status. Given the limited data, observation period and the need for further investigation, the influence of drying and wetting cycles on organic matter degradation and on  $^{137}\text{Cs}$  availability was not really clear.

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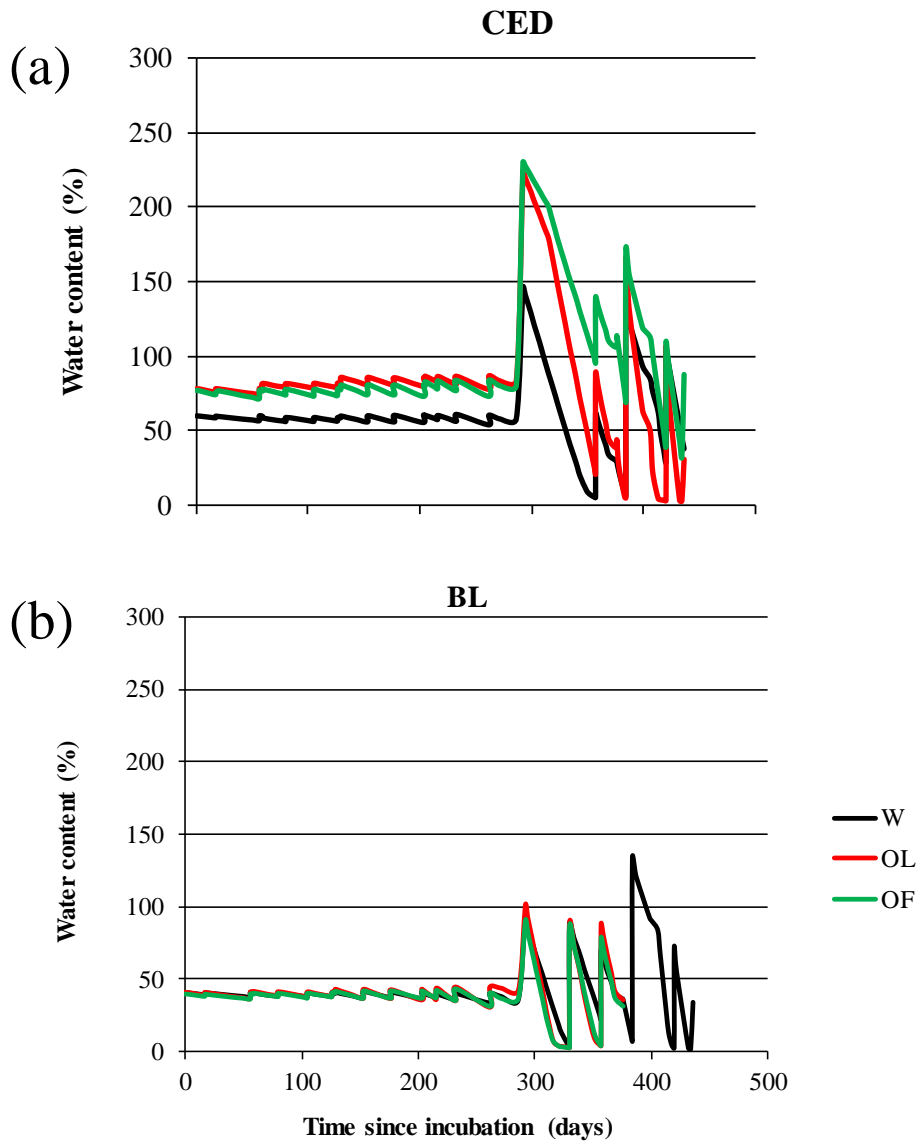
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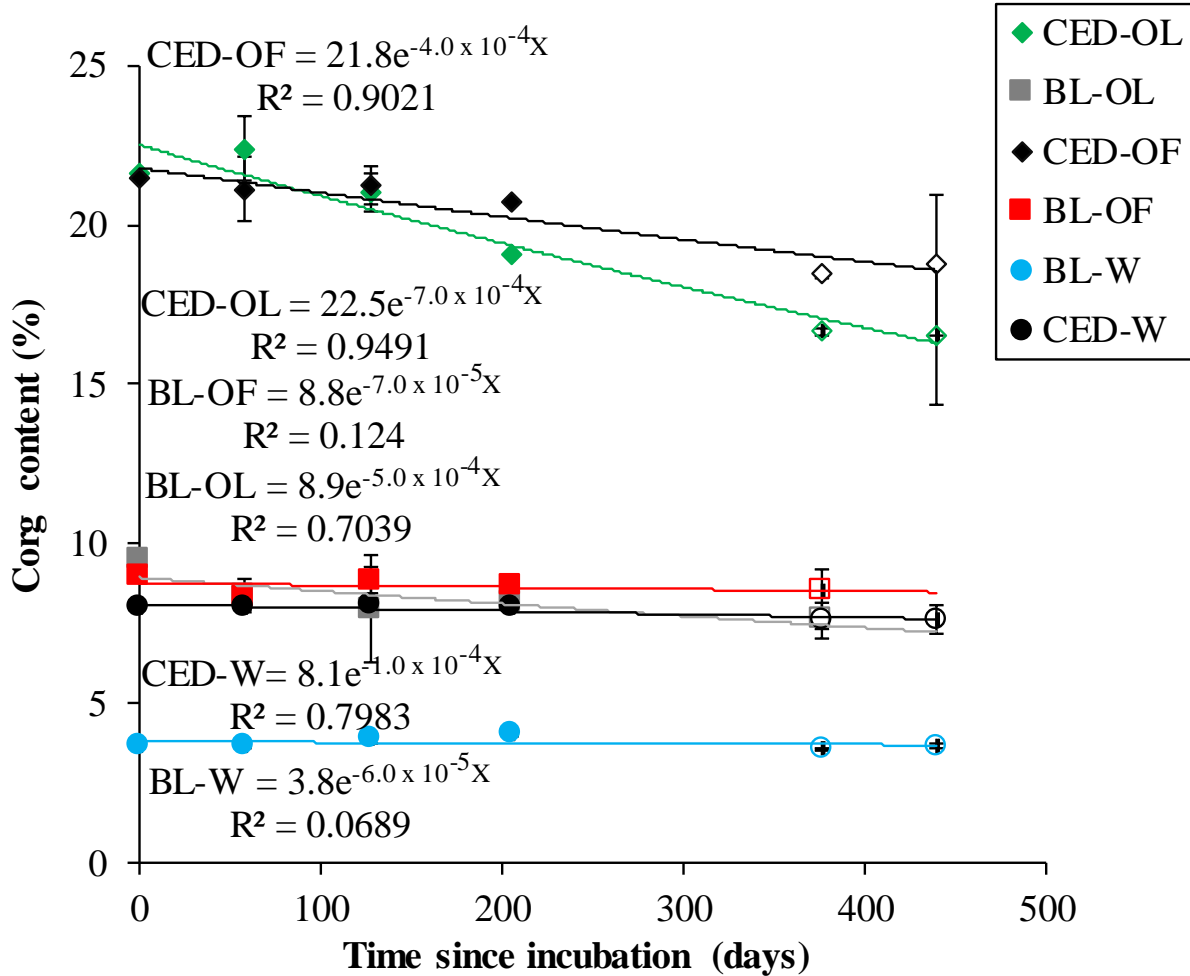
**Table 1.** The evolution of the extractable fraction of  $^{137}\text{Cs}$  and its  $k'_d$  with respect to the incubation time. The shaded cells represent the values obtained after the third (at t-377) and fourth (at t-440) complete drying and wetting cycles. ( $n = 2$ )

Forest type	Time (d)	Extracted by water		Extracted by acetate					
		$^{137}\text{Cs}$ liquid contamination		$^{137}\text{Cs}$ liquid contamination		$^{137}\text{Cs}$ solid contamination			
		W $k'_d$ (L kg $^{-1}$ )	Extractable %	W $k'_d$ (L kg $^{-1}$ )	Extractable %	OL $k'_d$ (L kg $^{-1}$ )	Extractable %	OF $k'_d$ (L kg $^{-1}$ )	Extractable %
BL-	t-0*	27.0±3.0	0.72±0.03	3.1±0.3	54.9±2.0	4.6±0.8	14.5±2.0	6.8±0.8	5.9±0.3
	t-59*	15.1±2.0	0.51±0.03	6.3±0.4	54.1±3.0	10.6±2.4	10.9±2.1	10.3±1.1	4.0±0.2
	t-129*	15.7±1.0	0.62±0.08	7.8±0.8	56.2±7.0	10.3±1.8	12.5±3.6	14.9±1.7	6.7±0.3
	t-205*	13.8±3.0	0.53±0.03	6.6±0.4	51.0±3.0	17.5±3.6	12.2±0.6	14.9±2.0	5.9±0.3
	t-377	12.1±0.6	0.43±0.02	6.9±0.4	43.8±2.2	21.7±8.0	17.0±5.3	14.1±2.2	6.3±0.5
	t-440	13.9±0.7	0.29±0.01	6.9±0.4	29.7±1.5				
CED-	t-0*	29.4±1.7	1.00±0.03	4.2±0.2	55.8±3.0	5.3±0.4	1.8±0.1	9.4±0.1	3.8±0.2
	t-59*	13.0±1.4	0.54±0.03	5.2±0.3	52.0±3.0	11.7±0.6	1.9±0.1	14.0±0.7	3.1±0.2
	t-129*	7.4±0.4	0.64±0.03	5.3±0.3	56.4±3.0	13.8±1.2	2.2±0.2	19.4±1.5	2.8±0.2
	t-205*	5.6±0.3	0.34±0.02	6.3±0.7	45.9±5.4	17.4±3.0	2.6±0.3	15.1±0.8	2.9±0.5
	t-377	5.4±0.4	0.20±0.03	6.5±0.3	36.2±2.3	16.8±0.8	4.9±1.9	20.8±1.0	4.2±0.2
	t-440	5.3±1.3	0.29±0.03	6.4±0.5	37.1±4.0	16.3±3.1	2.1±0.1	16.8±1.0	3.9±0.2

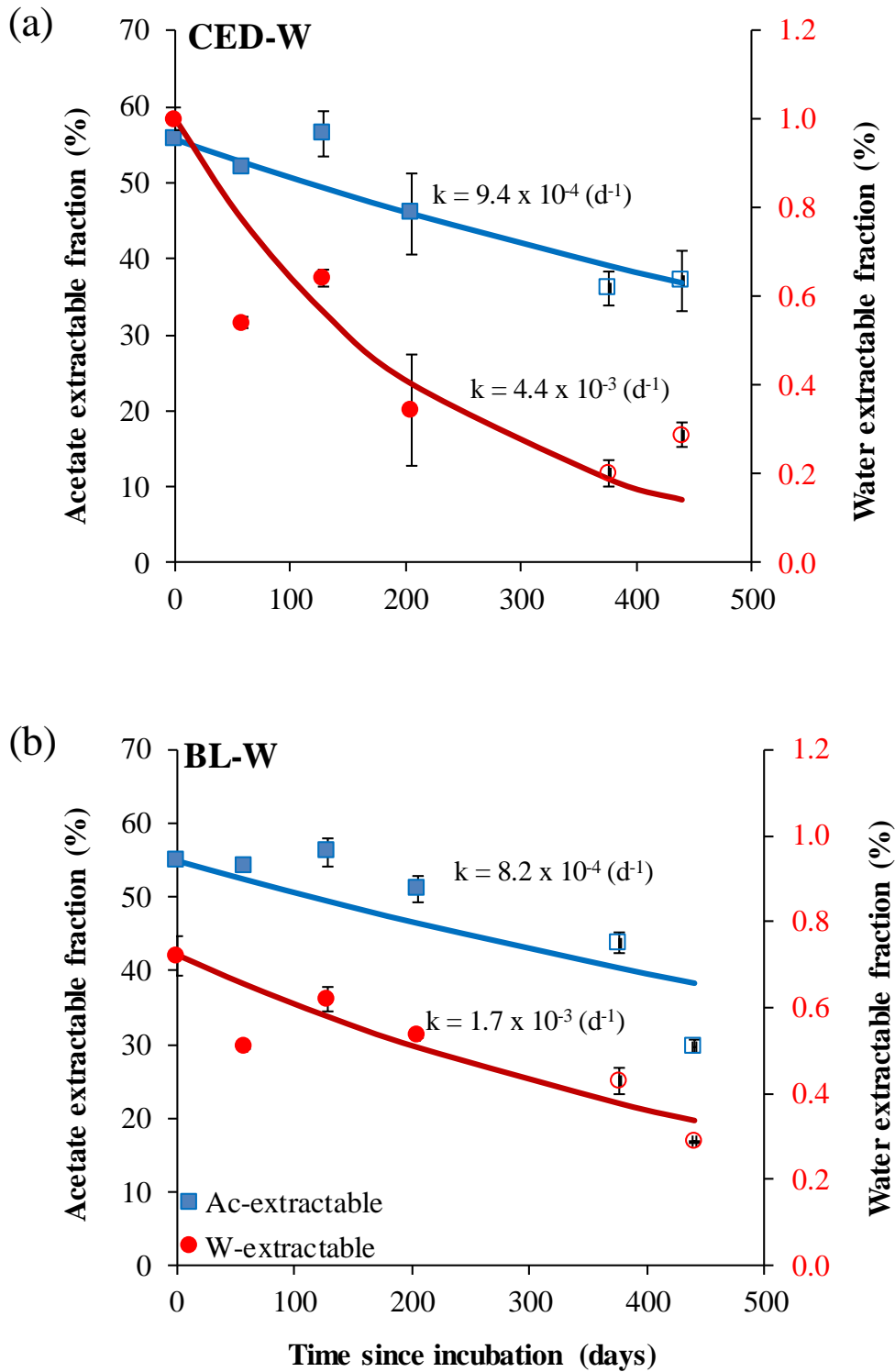
Note: -\*: data obtained from [Teramage et al. \(2018\)](#); BL: Broadleaf forest; CED: Cedar forest; W: water/aqueous/liquid; OL: litter; OF: fragmented litter, mean ± standard deviation.



**Figure 1.** The moisture regime of the soil during incubation period for liquid contamination (W), added contaminated litter (OL) and added contaminated fragmented litter material (OF).



**Figure 2.** Evolution of  $C_{org}$  content in the soils contaminated with liquid (BL-W, CED-W) or with organic materials  $^{137}Cs$  inputs (BL-OL, BL-OF, CED-OL, CED-OF). The non-filled marks represent the data during drying and wetting treatments and filled marks represents during constant moisture content.



**Figure 3.** Acetate- and water-extractable  $^{137}\text{Cs}$  fractions and their respective kinetics of stabilization for soils contaminated with liquid  $^{137}\text{Cs}$  input (CED-W and BL-W). Filled and non-filled marks represent the data before and after drying and wetting cycles, respectively.