

Vertical distributions of radiocesium in Japanese forest soils following the Fukushima Daiichi Nuclear Power Plant accident: A meta-analysis

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ABSTRACT

This study investigated the temporal change in vertical distributions of radiocesium inventories in Japanese forest soils during the early phase (from 2011 to 2017) following the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, using three simple parameters. We calculated the fraction in the organic layer ($F_{l/t}$), the migration center (X_c) and the relaxation depth (α) using 99 soil inventory data sets. $F_{l/t}$ decreased significantly from 2011 to 2017 (logistic analysis, $p < 0.001$). In addition, $F_{l/t}$ in the FDNPP zone rapidly decreased compared to that in the Chernobyl Nuclear Power Plant (ChNPP) zone from the first year to the second year. Different migration rates from organic to mineral soil layers between previous studies in the ChNPP and this study have several possible causes such as organic litter features, climate and physico-chemical forms of initial deposition. In mineral soil layers in the FDNPP zone, only X_c increased significantly with time according to generalized mixed model analysis ($p < 0.01$). However, X_c and α in the ChNPP zone decreased from two to five years after the accident in 1986, which shows a high ^{137}Cs retention in the organic layer even in the fifth year after the accident. The vertical migration of ^{137}Cs in the mineral soil layer in the FDNPP zone appears to be due to low input of ^{137}Cs from organic to surface mineral soil layer after the second year. These results indicate that ^{137}Cs retention capacity of the organic layer can affect the apparent vertical migration of ^{137}Cs in the underlying mineral soil layer.

1. Introduction

Radiocesium emitted to the atmosphere from the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident deposited on extensive forest areas of East Japan in March 2011 (Hashimoto et al., 2012; Kato et al., 2019). Previous studies have reported that radiocesium that deposited on forest ecosystems after the Chernobyl Nuclear Power Plant (ChNPP) accident became integrated in the natural elemental cycles of the forests (Tikhomirov and Shcheglov, 1994; Shcheglov et al., 2001; IAEA, 2002; Goor and Thiry, 2004). A varying portion of radiocesium that deposited directly on the forest canopy was absorbed by leaves and needles (Myttenaere et al., 1993; Nimis, 1996; Thiry et al., 2016). The majority of radiocesium deposits was rapidly transferred to the forest floor after

removal from the canopy by precipitation and loss of leaves or needles (Pröhl, 2009; Bunzl et al., 1989; Bonnett and Anderson, 1993). With time, this radiocesium migrated downwards into the underlying mineral soil where it is partly available for root uptake by trees and understory vegetation (Fesenko et al., 2001a,b). Modeling studies have shown that radiocesium cycling in forest ecosystems, particularly the transfer from tree canopies to the forest floor, is very dynamic in the early phase after atmospheric fallout (Nishina et al., 2018; Thiry et al., 2018; Hashimoto et al., 2020). The influence of soil conditions on root uptake increases with time (Thiry et al., 2020). Therefore, quantitative understanding of the migration of radiocesium in forest soils, the primary source of long-term tree contamination, is necessary to forecast radiocesium cycling in forest ecosystems in the coming decades. This information is

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Table 1
Site information of previous Chernobyl studies.

No.	Site name	Distance from ChNPP (km)	¹³⁷ Cs deposition (kBq m ⁻²)	Dry or wet deposition	Mean annual precipitation (mm)	Mean annual temperature (°C)	Tree species	Litter weight (kg m ⁻²)	Litter thickness (cm)	Soil type	Research period	Reference
1	Kruki, Belarus	25	6646 ^a	Dry deposition	652 ^b	7.9 ^b	Scots pine	–	5	Soddy podzolic soil	1992–1997	Belli et al. (2000); IAEA (2006) Shcheglov et al. (2001)
2	Dityatki-1, Ukraine	28.5	240	Dry deposition	611 ^c	8.4 ^c	Mixed, broad-leaved-pine	–	6	Podzolic, iron-illuvial, sandy soil	1986–1999	IAEA (2002, 2006, Table 1)
	Kopachi-2, Ukraine	6.5	2900	Dry deposition			Pine	7	5	Secondary-podzolic, sandy soil	1986–1999	
	Shepelichi-1, Ukraine	6	44,730	Dry deposition			Mixed, broad-leaved-pine	–	4	Weak-podzolic, weakly stratified, sandy soil	1986–1999	
	Dityatki-3, Ukraine	26	240	Dry deposition			Alder	–	5	Bog, valley peat-gley, soil	1986–1999	
3	Zhitomir, Ukraine	130	555	Dry deposition	550–710	7.9	Pine with sparse birch	4.4	8	Soddy podzolic sandy soil	1991–1997	IAEA (2002)

^a Thiry et al. (2020).

^b Mean annual value at Homiel, Belarus (Japan Meteorological Agency).

^c Mean annual value at Kiev, Ukraine (Japan Meteorological Agency).

also important to understand the potential supply of radioactive contamination stored in forest areas to downstream ecosystems (Kurikami et al., 2019; Laceby et al., 2016) because forest ecosystems have not been decontaminated in the FDNPP-impacted area.

After the ChNPP accident, a few studies reported the vertical distribution of radiocesium in forest soils over a period of several years (Belli, 2000; Shcheglov et al., 2001; IAEA, 2002, 2006, Table 1). These previous studies all showed distinct changes in vertical migration of radiocesium in forest soils in the ChNPP zone over several years. After the FDNPP accident, numerous studies have reported the vertical distribution of radiocesium in a variety of Japanese forest soils (Table 2). However, many studies after the FDNPP accident measured vertical distribution of ¹³⁷Cs during one single sampling campaign. The number of studies which have monitored radiocesium migration for several years after the FDNPP accident is limited (Imamura et al., 2017; Kinno et al., 2017; Takahashi et al., 2018; Muto et al., 2019). Imamura et al. (2017) reported vertical distributions of ¹³⁷Cs activity inventories at five forest sites from 2011 to 2015. Kinno et al. (2017) showed vertical distributions of ¹³⁷Cs activity concentrations in deciduous and Japanese cedar forests in Namie Town from 2011 to 2016. Takahashi et al. (2018) reported the vertical distributions of ¹³⁷Cs activity inventories in three forests in Kawamata Town from 2011 to 2016. Muto et al. (2019) reported time-series changes in vertical distribution of ¹³⁷Cs activity inventories in Fukushima City from 2011 to 2015. The cesium retention capacities of soils vary between individual sites depending on climate, land use and soil types (Rosén et al., 1999). Because factors such as the physico-chemical form of the initial deposition, temperature and precipitation, litter thickness and soil types are different between the FDNPP and Chernobyl accident zones, changes in vertical distributions of radiocesium with time after deposition could be very different in these two areas.

In this paper we describe the change in vertical distributions of ¹³⁷Cs in forest soils during the early phase following the FDNPP accident by gathering observation results in one single year and several year samplings, all based on soil inventory data from 99 soils studied from 2011 to 2017 after the FDNPP accident. Migration of radiocesium through

organic and mineral soil layers is described using meta-data analysis of time series. These analyses are summarised using three simple parameters: (i) the fraction of total deposition remaining in the organic layer (F_{lv}), (ii) the migration center (X_c) and (iii) the relaxation depth (α). The parameters obtained from meta-data analysis can be used to predict ¹³⁷Cs dynamics in the FDNPP zone and to compare these with observations in forest soils in the ChNPP zone.

2. Materials and methods

2.1. Data description

A meta-analysis was conducted based on 99 vertical soil profiles of radiocesium presented in six publications (Table 3). These publications were identified using three words “Fukushima”, “forest”, and “cesium” in Web of Science during 2011 and 2017 and soil profiles of ¹³⁷Cs activity inventory data extracted from them. 76% of study sites were located in the area adjacent to the FDNPP with over 30 kBq m⁻² total cesium deposition (Fig. 1). ¹³⁷Cs was deposited by dry deposition only near the FDNPP area (Terada et al., 2012). In addition, some studies reported that ¹³⁷Cs deposited in the form of small particles (‘cesium balls’) (Adachi et al., 2013; Yamaguchi et al., 2016). Climate in the FDNPP area is temperate. The data sets cover forests comprising four tree species: Japanese cedar (*Cryptomeria japonica*), Hinoki cypress (*Chamaecyparis obtuse*), Konara oak (*Quercus serrata*) and Red pine (*Pinus densiflora*). Litter layer thicknesses at these sites ranged from 0 to 6 cm. The soil types were mainly Andosols (58% of study sites) but there were also Fluvisols and/or Cambisols (brown forest soils). The deepest soil sampling depth in most of the studies was 20 cm but soil sampling depth intervals were different between the studies (Table S1).

Three long-term monitoring data sets were found mainly for the 30 km exclusion zone in the ChNPP zone (Belli, 2000; Shcheglov et al., 2001; IAEA, 2002, 2006, Table 1). Radiocesium in the 30-km zone deposited as dry, finely-dispersed particles or larger hot particles (Tikhomirov and Shcheglov, 1994; Shcheglov et al., 2001). Climate in the ChNPP area is microthermal. Litter layer thicknesses were from 3 to

Table 2
Studies reported the vertical distribution of radiocesium in a variety of Japanese forest soil.

No.	Distance from FDNPP (km)	Location	Latitude (°)	Longitude (°)	Study period	Sampling depth and interval (cm)	Reference
1	70	Fukushima, Fukushima	37.71	140.36	June 2011	0–1, 1–3, 3–5, 5–10, >10	Koarashi et al. (2012)
2	60	Koriyama, Fukushima	37.48	140.39	April 2011	0–2, 2–4, 4–6, 6–10, 10–15	Ohno et al. (2012)
3	195	Kashiwa, Chiba	35.90	139.93	September 2011	0–2, 2–3, 3–4, 4–5, 5–6, 6–8, 8–10, 10–12, 12–14. 0–1, 1–2, 2–3, 3–4, 4–5, 5–6, 6–8, 8–10	Fukuda et al. (2013)
4	70	Fukushima, Fukushima	37.71	140.36	July 2011	0–1, 1–3, 3–5, 5–10, >10	Matsunaga et al. (2013)
5	60	Koriyama, Fukushima	37.38	140.25	June 2013	0–0.5, 0.5–3, 3–8, 8–12, 12–18, 18–25, 25–35, 35–45	Mahara et al. (2014)
6	30	Namie, Fukushima	37.56	140.75	April 2013	0–0.5, 0.5–1, 1–1.5, 1.5–2, 2–2.5, 2.5–3, 3–3.5, 3.5–4, 4–4.5, 4.5–5, 5–6, 6–7, 7–8, 8–9, 9–10	Kuroshima et al. (2014)
7	59	Fukushima, Fukushima	37.68	140.45	August 2013	0–1, 1–3, 3–5, 5–7, 7–9, 9–11, 11–14, 14–18, 18–23, 23–28	Fujiyoshi et al. (2015)
8	18, 16	Kawauchi, Fukushima	37.34, 37.34	140.85, 140.88	September 2012	0–1, 1–2, 2–3, 3–4, 4–5, 5–6, 6–7, 7–8, 8–9, 9–10, 10–11, 11–12, 12–13, 13–14, 14–15, 15–16, 16–17, 17–18, 18–19, 19–20	Nakai et al. (2015)
9	160	Ishioka, Ibaraki	36.20	140.13	April 2011	0–2, 2–4, 4–6	Nishikiori et al. (2015)
10	26	Kawauchi, Fukushima	37.29	140.80	August 2011	0–5, 5–10, 10–15, 15–20	Komatsu et al. (2015)
	66	Otama, Fukushima	37.58	140.31	August 2011	0–5, 5–10, 10–15, 15–20	
	134	Tadami, Fukushima	37.32	139.52	September 2011	0–5, 5–10, 10–15, 15–20	
11	40	Kawamata, Fukushima	37.58	140.69	November 2013	0–3, 3–8, 8–20	Coppin et al. (2016)
12	7	Okuma, Fukushima	37.38	140.94	March 2014	0–2, 2–5, 5–10, 10–20, 20–28.5	Konoplev et al. (2016)
	8	Okuma, Fukushima	37.38	140.94	March 2014	0–5, 5–10, 10–15, 15–21	
13		Namie, Fukushima	37.42	141.01	November 2012	0–2, 2–4, 4–6, 6–8, 8–10, 10–15, 15–20	Mishra et al. (2016)
		Namie, Fukushima	37.42	141.01	September 2012	0–2, 2–4, 4–6, 6–8, 8–13	
	23	Namie, Fukushima	37.55	140.84	November 2012	0–2, 2–4, 4–6, 6–8, 8–10, 10–15, 15–20	
	23	Namie, Fukushima	37.55	140.83	June 2013	0–5, 5–10, 10–15, 15–20	
	28	Futaba, Fukushima	37.55	140.75	June 2013	0–5, 5–10, 10–15, 15–20	
14	37	Kawamoto, Fukushima	37.59	140.69	October 2014	0–5, 5–10, 10–15, 15–20	Sugiura et al. (2016)
15	180	Sano, Tochigi	36.39	139.74	January, March, May, October 2012	0–0.5, 0.5–1, 1–1.5, 1.5–2, 2–2.5, 2.5–3, 3–3.5, 3.5–4, 4–4.5, 4.5–5, 5–6, 6–7, 7–8, 8–9, 9–10, 10–12, 12–14, 14–16, 16–18, 18–20, 20–22, 22–24, 24–26, 26–28, 28–30	Teramage et al. (2016)
16	52	Date, Fukushima	37.73	140.58	March 2013	0–4, 4–8, 8–12, 12–16, 16–20, 20–22	Xu et al. (2016)
						0–3, 3–6, 6–9, 9–12, 12–15, 15–17	
17	250	Morioka, Iwate	39.78	141.16	October 2012	0–2, 2–4, 4–6, 6–8, 8–10, 10–15, 15–20, 20–30	Kang et al. (2017)

8 cm. The soil types were mainly Podzols, developing in automorphic (i.e., dry) conditions. One site (Dityatki-3) was, however, characterised by hydromorphic (i.e., wet) soil conditions. In addition, the site at Zhitomir, about 130 km to south-west of Chernobyl, was characterised by semi-hydromorphic soil conditions. At all sites, Scots pine (*Pinus sylvestris* L.) was the predominant tree species.

2.2. Data analysis

2.2.1. Fraction in the organic layer

The fraction of radiocesium retained in the organic layer was calculated to describe the time series of migration of ¹³⁷Cs activity inventories from organic layers to mineral soil layers. The ratio of the radiocesium activity inventory in the organic layer to the sum of that in the organic layer and mineral soil layers is given by:

$$F_{l/t} = \frac{I_l}{I_l + I_s} \times 100 (\%) \tag{1}$$

where $F_{l/t}$ is the fraction in the organic layer, I_l is the inventory in the organic layer and I_s is the inventory in the mineral soil layer.

2.2.2. Migration center

The migration center is the depth at which the center of gravity of ¹³⁷Cs is located in the soil profile; this was calculated using the following expression (Arapis et al., 1997; Ramzaev and Barkovsky, 2018; Fujii et al., 2019).

$$X_c = \frac{\sum_i X_i I_i}{\sum_i I_i} \tag{2}$$

where X_c is the migration center (cm), X_i is the center of the measured layer i (cm) and I_i is the ¹³⁷Cs activity inventory (Bq m⁻²) within the measured layer i .

2.2.3. Relaxation depth

The ¹³⁷Cs activity inventory decreased exponentially with depth in

Table 3

Site Information of data in six publications after FDNPP accident.

No.	Site name	Latitude (°)	Longitude (°)	Distance from FDNPP (km)	¹³⁷ Cs deposition (kBq m ⁻²) ^a	Dry or wet deposition ^b	Mean annual precipitation (mm) ^c	Mean annual temperature (°C)	Tree species	Litter weight (kg m ⁻²)	Litter layer thickness (cm)	Soil type	Research period	Reference						
1	FR-1	37.71	140.36	78	28	Wet	1365.7	8.1	Oak	n.a.	–	Fluvisols	June 2011	Koarashi et al. (2012)						
	FR-2								Oak	n.a.	–	Fluvisols								
	FR-3								Pine	n.a.	–	Fluvisols								
	FR-4								Japanese cedar	n.a.	–	Andosols								
	FR-5								Japanese cedar	n.a.	–	Andosols								
2	FR-1	37.71	140.36	68	37	Wet	1701.7	11.2	Oak	n.a.	–	Fluvisols	July 2011	Matsunaga et al. (2013)						
	FR-2								Oak	n.a.	–	Fluvisols								
	FR-3								Pine	n.a.	–	Fluvisols								
	FR-4								Japanese cedar	n.a.	–	Andosols								
	FR-5								Japanese cedar	n.a.	–	Andosols								
3	KAW	37.58	140.68	36	502	Wet	1205.7	9.1	Konara oak, Maple	–	5	Cambisols (brown forest soils)	October 2014	Sugiura et al. (2016)						
4	KU1-S	37.29	140.80	26	535	Dry	1445.8	8.9	Japanese cedar	1.1–1.6	+2–0	Andosols or Cambisols (brown forest soils)	2011–2015	Imamura et al. (2017)						
	KU1-H								140.79	26	535	1431.7			8.4	Hinoki cypress	1.1–1.5	–	–	
	KU1-Q	37.29	140.79	26	535	1431.7	8.4	Konara oak	1.0–1.2	–	–									
	KU2-S	37.38	140.72	28	159	Dry	1351.6	8	Japanese cedar	1.4–2.8	–	–								
	OT-S	37.58	140.31	67	42	Wet	1752.5	8.2	Japanese cedar	1.6–2.1	+2–0	Andosols or black soils								
	OT-Q								37.57	140.31	66	42	1752.5	8.2	Konara oak	0.9–1.5	+2–0	Andosols or Cambisols (brown forest soils)		
	OT-P								37.57	140.31	66	42	1752.5	8.2	Red pine	1.0–1.7	+2–0	Andosols or Cambisols (brown forest soils)		
TD-S	37.32	139.52	135	5	Wet	1506.0	8.4	Japanese cedar	1.1–1.6	+2–0	Inceptisols or Cambisols (brown forest soils)									
TB-H	36.17	140.18	158	18.5	Wet	1235.8	12.3	Hinoki cypress	0.8–1.5	–	–									
5	CF-1-3	39.77	141.15	250	<10	Wet	1240.4	9.3	Japanese cedar	–	1–2	Andosols	October 2012	Kang et al. (2017)						
	CW-1-3								–	1–2	Andosols									
	CS-1-3								–	1–2	Andosols									
	OF-1-3								Konara oak	–	1–2	Andosols								
	OW-1-3								–	1–2	Andosols									
OS-1-3	–	1–2	Andosols																	
6	MF	37.60	140.68	37	412	Wet and Dry	1196.9	9.3	Konara oak, Red pine	1.84	+0–6	Aluandic Andosol	2011–2016	Takahashi et al. (2018)						
	MC								37.59	140.69	35	483			1212.9	9.1	Japanese cedar	2.75	+0–5	Aluandic Andosol
	YC																37.59	140.69	35	483

^a Air born monitoring on 28 June 2012 (MEXT, 2012).

^b Calculated by Fig. 9c of Terada et al. (2012).

^c National Land Numerical Information.

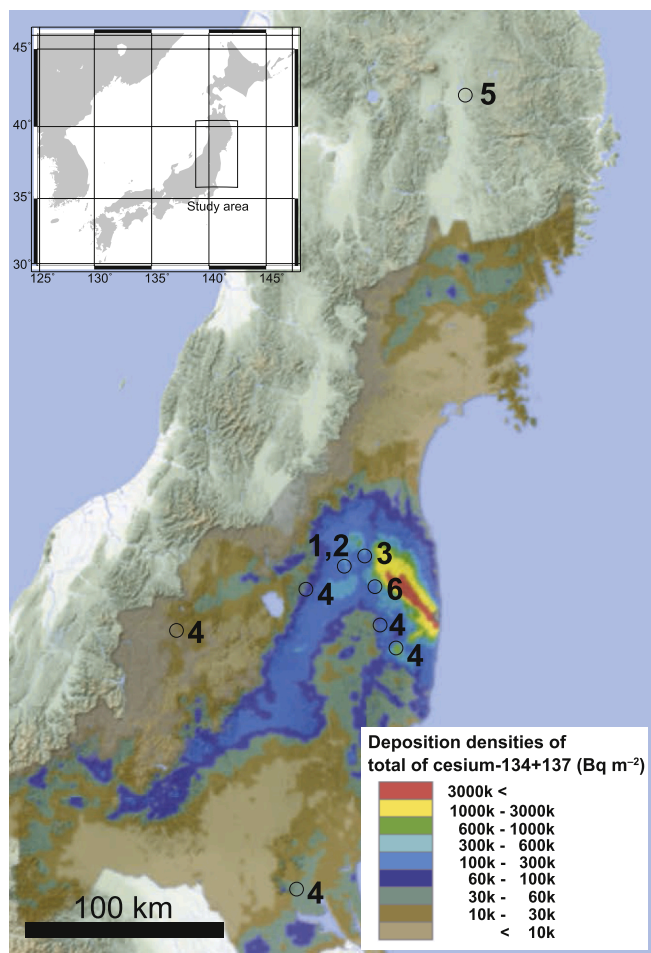


Fig. 1. Location of study sites. 1: Koarashi et al. (2012), 2: Matsunaga et al. (2013), 3: Sugiura et al. (2016), 4: Imamura et al. (2017), 5: Kang et al. (2017), 6: Takahashi et al. (2018). Map with the ¹³⁴Cs and ¹³⁷Cs deposition levels on June 28, 2012, generated by using the website “Extension Site of Distribution Map of Radiation Dose, etc.” (MEXT, 2012).

most soil profiles (Table S1). Therefore, we used the relaxation depth to quantify the exponential migration of ¹³⁷Cs activity inventory in the mineral soil layer (Kato et al., 2012; Koarashi et al., 2012; Matsunaga et al., 2013). The depth at which the radiocesium activity inventory at the soil surface ($I(0)$) decreases to $1/e$ (approximately 37%):

$$I(x) = R \left(1 - e^{-\frac{x}{\alpha}} \right) \quad (3)$$

where $I(x)$ is the cumulative ¹³⁷Cs activity inventory (Bq m^{-2}) down to depth x (cm), R is the total ¹³⁷Cs activity inventory in mineral soil (Bq m^{-2}) and α is the relaxation depth (cm). x is the lowermost depth of each cumulative mineral soil layer.

2.2.4. Long-term analysis

Time-series changes of the three simple parameters ($F_{l/t}$, X_c and α ; Table S2) in the long-term after the FDNPP accident were analysed. The statistical significance of time-series trends for each parameter was tested using mixed-effects models accounting for sampling sites as a random effect, because the three parameters differed at each sampling site. Linear and exponential regression models were used for all parameters, and logistic regression model was fitted to $F_{l/t}$. The Akaike information criterion (AIC) of all models was calculated including fixed-effects models and the model with the smallest AIC value was adopted for each parameter. These statistical analyses were performed using R, version 3.6.3 (R Development Core Team, 2020).

3. Results and discussions

3.1. Migration of ¹³⁷Cs from organic to mineral soil layers

The time series of $F_{l/t}$ is shown in Fig. 2. $F_{l/t}$ decreased significantly from 2011 to 2017 (mixed-effects logistic regression model, $p < 0.001$) in forest areas affected by Fukushima fallout. AIC values of mixed models were clearly lower than these of fixed models. This indicated that radiocesium gradually migrated from organic to mineral soil layers during the early phase (from 2011 to 2017) following the accident, although the fraction of radiocesium in an organic layer is greatly affected by the difference of sampling site (median $F_{l/t}$ = 60% and 6% in 2011 and 2017, respectively). To compare ¹³⁷Cs migration in forest soils in the FDNPP and ChNPP zones, $F_{l/t}$ in the first, second and fifth years following each accident are shown in Fig. 3. Although there is a large range of variation in FDNPP, this shows that $F_{l/t}$ in the FDNPP zone decreased much more rapidly, especially from the first year to the second year, compared to that in the ChNPP zone. From the first to the second year after deposition, the median $F_{l/t}$ decreased by 32% in soils in the FDNPP but only by 3% in the ChNPP zone. Koarashi and Atarashi-Andoh (2019) and Muto et al. (2019) compared retention capacity of the organic layer between European and Japanese forest ecosystems. Their results agreed with our results which compared FDNPP with ChNPP zone. Shcheglov et al. (2001) reported the rate of vertical distributions of ¹³⁷Cs activity inventories at four forests in Ukraine from 1986 to 1999, showing significant movement of ¹³⁷Cs from the A01 layer

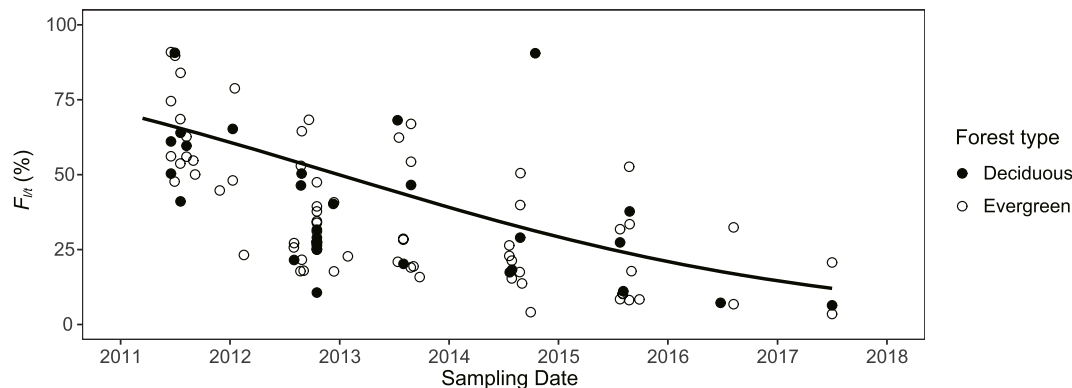


Fig. 2. Time series of the fraction of radiocesium retained in the organic layer ($F_{l/t}$) in FDNPP forest soils. A significantly negative temporal trend was observed using a mixed-effects logistic regression model (solid line, $p < 0.001$).

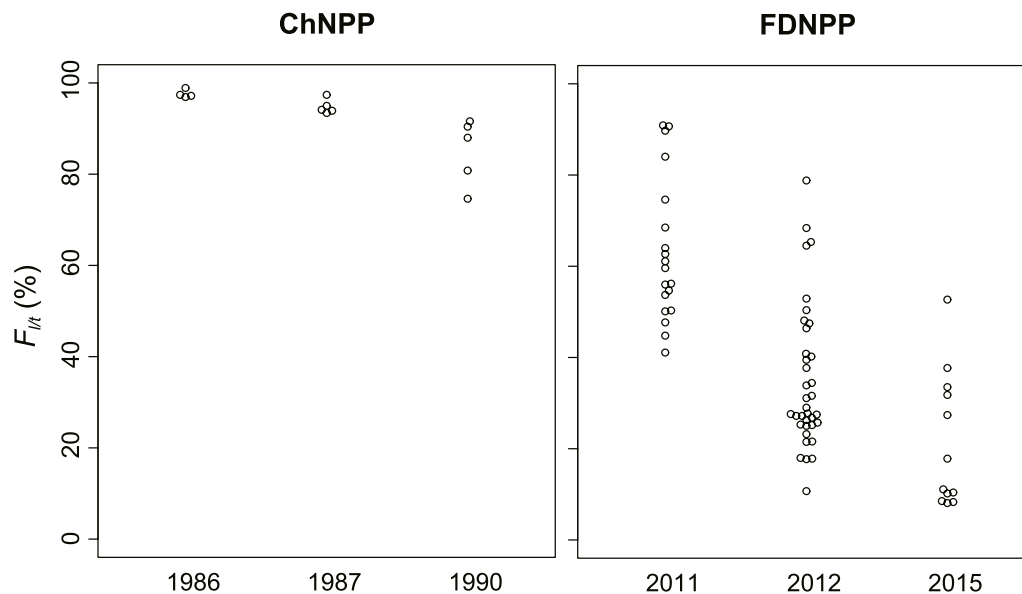


Fig. 3. F_{lt} in first, second and fifth years after the both accident (ChNPP and FDNPP).

to the A0f and A0h layers in the organic layer from 1986 to 1987; approximately 90% of ^{137}Cs was retained in the A0l layer in 1986, but in 1987 this reduced to about 35%–50%, with approximately 30%–40% and 10% in A0f and A0h layers, respectively. This indicates that ^{137}Cs did not move rapidly to the mineral soil layer from the organic layer within one year after the accident in these Ukrainian forests. In addition, F_{lt} in the first year in the ChNPP zone was much higher (median $F_{lt} = 97\%$) than in the FDNPP zone (median $F_{lt} = 60\%$). ^{137}Cs was not as readily leached from the organic layer by rainfall in forest soils in the ChNPP zone as it was in the FDNPP forests, possibly because of differences in the physico-chemical form of the initial deposition at the two accident sites. ^{137}Cs contamination in the ChNPP zone was dominated by dry deposition of small or large particles (Tikhomirov and Shcheglov, 1994; Shcheglov et al., 2001) while in forests affected by FDNPP fallout, wet deposition in addition to throughfall was the predominant form of ^{137}Cs transferred to soil surfaces in the first year because almost all sites were affected by wet deposition in this study (except KU1 and KU2 in No.4; Table 3 and Terada et al., 2012). In fact, because there was only a minor difference in vertical distribution of ^{137}Cs before and after the rain event in July 2011 (Matsunaga et al., 2013), radiocesium must have moved to the surface mineral soil layer from the organic layer immediately after the accident in the FDNPP zone. It is thus possible that ^{137}Cs migration was more significantly influenced by preferential water flows leading to a rapid initial redistribution in surface soil layers in a large

proportion of Japanese contaminated forest.

Between two and five years after each respective accident (Fig. 3), the decrease in F_{lt} in forest soils in the Chernobyl zone (6% reduction in median F_{lt}) was lower than in the FDNPP zone (14% reduction in median F_{lt}). In the long-term, radiocesium accumulation and persistence in the organic layer depends on litter weight and/or litter layer thickness (Fesenko et al., 2001a; Shcheglov et al., 2001; Goor et al., 2007; Winkelbauer et al., 2012; Koarashi et al., 2016; Ito et al., 2018). Litter weight in Chernobyl and Japanese forest soils were reported to be $4.4\text{--}7.0\text{ kg m}^{-2}$ and $0.8\text{--}3.0\text{ kg m}^{-2}$, respectively (Tables 1 and 3). The thickness of organic layers in ChNPP and Japanese forest soils were listed to be 3–8 cm and 0–6 cm, respectively (Tables 1 and 3). In general, the organic layers in the Japanese forest soils were lighter and thinner compared to those in the ChNPP zone and there was no A0h layer in Japanese forests (Takahashi et al., 2018). That feature can facilitate the vertical migration of ^{137}Cs . Radiocesium movement from the organic layer to the mineral soil layer is also affected by leaching due to precipitation. Whereas mean annual precipitation in the ChNPP zone is about 600 mm, it is much higher in Japanese forests, ranging from 1200 mm to 1700 mm (Tables 1 and 3). In addition, the mean annual temperature in Japan is higher than in Ukraine and Belarus (Tables 1 and 3). Therefore, weather conditions are also more favorable in Japan for a higher ^{137}Cs migration out of the forest floor through leaching or decomposition of organic material. The differences observed in the migration rate of

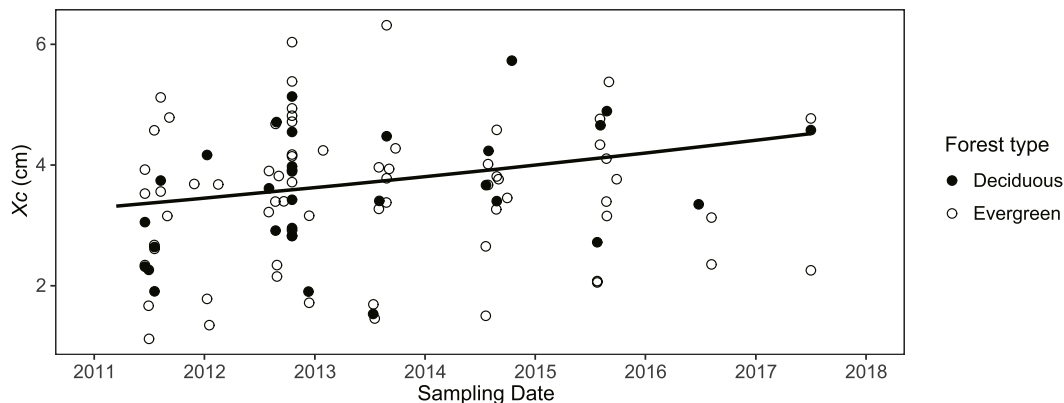


Fig. 4. Time series of migration center (X_c) in FDNPP forest soils. A positive temporal trend was indicated using a mixed-effects exponential regression model ($p < 0.01$).

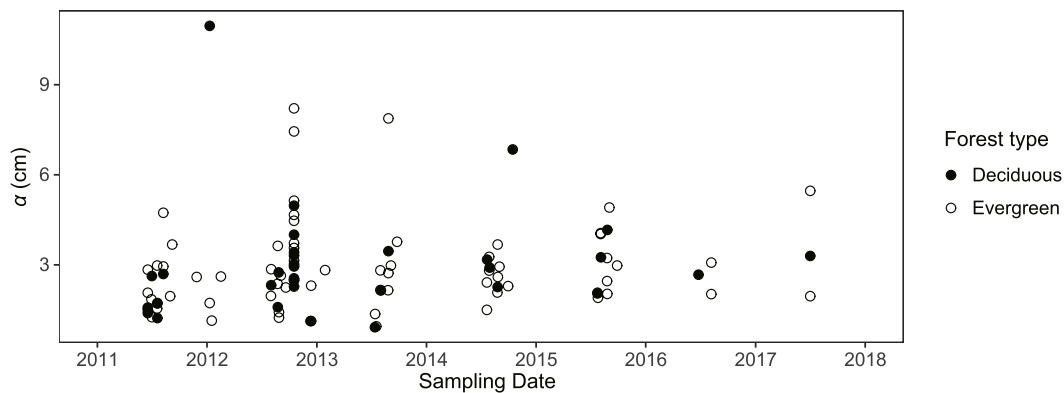


Fig. 5. Time series of relaxation depth (α) in FDNPP forest soils. The time-series trend was not significant according to a mixed-effects regression analysis ($p = 0.21$).

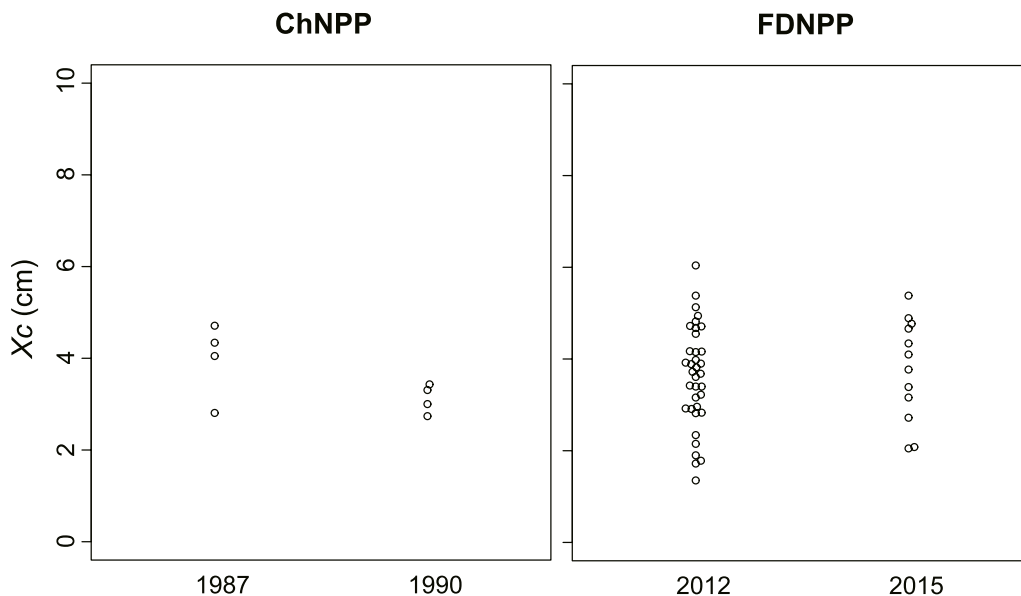


Fig. 6. Migration center (X_c) in second and five years after the both accident (ChNPP and FDNPP).

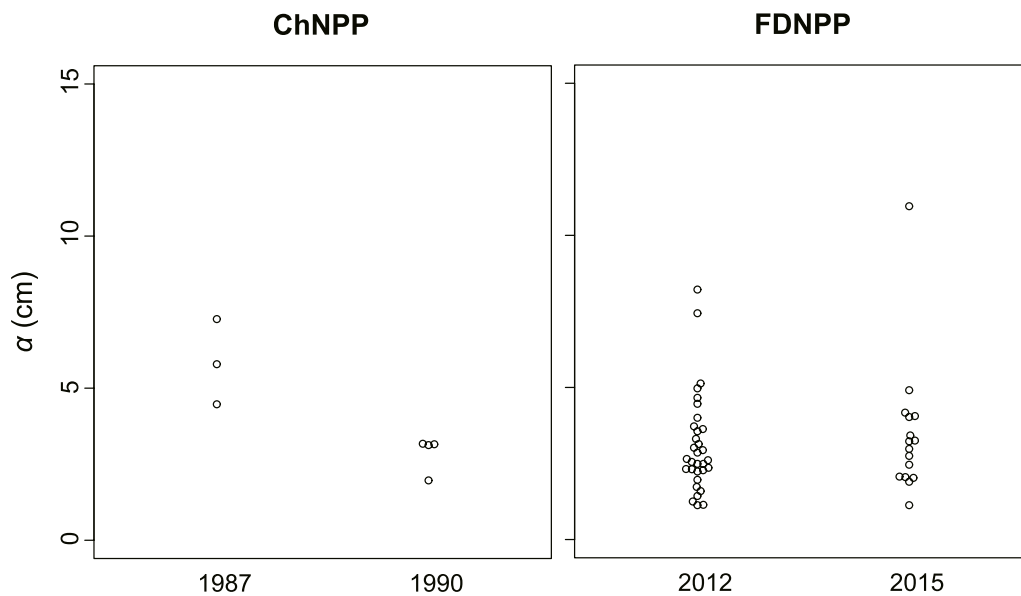


Fig. 7. Relaxation depth (α) in second and five years after the both accident (ChNPP and FDNPP).

radiocesium from organic to mineral soil layers between the Chernobyl and Japanese forest areas are likely to be due to differences in various factors such as edaphic and weather conditions, in addition to the form of the initial deposit.

3.2. Migration of ^{137}Cs in the mineral soil layer

Time series of X_c and α are shown in Figs. 4 and 5, respectively. Whereas α did not show significant change with time (mixed-effects linear regression model, $p = 0.21$), X_c significantly increased with time (mixed-effects exponential regression model, $p < 0.01$). Parameter α did not show significant change with time due to some outlier (Table S2). However, α also showed the same increasing trend as X_c . These results indicate that ^{137}Cs migrated from the upper to the deeper layers of the mineral soil during the early phase following the FDNPP accident because X_c showed a significant positive change. In contrast, both X_c and α decreased over this period in the ChNPP accident zone (1987 and 1990, Figs. 6 and 7). These results indicate that apparent vertical migration of ^{137}Cs with depth was observed in mineral soil only in the FDNPP area. In the ChNPP zone, ^{137}Cs activity inventories in the organic layers were over 7 times higher than in the surface mineral soils even five years after the accident (except at hydromorphic sites), suggesting the slow Cs migration to surface mineral soils from organic layers. In contrast, in the FDNPP zone, ^{137}Cs activity inventories in the organic layer were lower than those in the surface mineral soils from two years after the accident at almost all sites (67% of all vertical soil profiles after 2013; Table S1). This indicated that apparent vertical migration of ^{137}Cs in mineral soil layer in the FDNPP zone was few influenced by the input of ^{137}Cs from organic to surface mineral soil layers after the second year of the accident. Muto et al. (2019) explained roughly comparative result of ^{137}Cs transfer in mineral soil with European forest studies by using a diffusion equation model. However, this is because ^{137}Cs inventory is higher in the organic layer than in surface mineral soil layers at many sites (76%) in Muto et al. (2019) similar to European forests.

We used three different parameters to quantify and summarise vertical migration of radiocesium in forest soils. However, values of X_c and α were influenced by the local sampling designs. Specifically, the depth of the soil profiles sampled and soil sampling intervals both affect estimates X_c and α . In addition, this research did not contain uncertainties for sampling and analysis for each reviewed data set because this information was not reported in the original references. Therefore, although we could not consider uncertainties to our analyses, we need to be cautious that the results of this parameter include these uncertainties. Furthermore, data in the ChNPP zone was limited which also indicates that caution is needed when comparing ^{137}Cs migration in soils of the ChNPP and FDNPP.

4. Conclusion

This study investigated the temporal change in the vertical distribution of radiocesium activity inventories in Japanese forest soils during the early phase (from 2011 to 2017) following the FDNPP accident using three parameters derived from a meta-analysis of 99 data sets. The results show that radiocesium gradually migrated from organic to mineral soil layers over time. After radiocesium had migrated from organic layers to the mineral soil beneath, further vertical migration occurred. We have shown that migration rates from organic to mineral soil layers and in mineral soil layers differ from those reported in ChNPP studies. This could be caused by multiple differences in edaphic and weather conditions between the two contaminated areas and, especially, differences in migration of ^{137}Cs from organic to mineral soil layers in Japanese forest soils and in the ChNPP zone.

Our observations are useful in evaluating past and future ambient beta/gamma dose rates and formulating decontamination policy in forests; they are also helpful in estimating the potential source depth for root uptake of trees and understory vegetation in modeling radionuclide

dynamics within forest ecosystems as a whole, as well as the risk of further contamination of downstream ecosystems due to run off.

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

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvrad.2020.106422>.

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