



Vertical distribution of ^{137}Cs in the contaminated soil of a spruce forest in Southern Germany 12 years after regeneration cutting



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ARTICLE INFO

Keywords:

^{137}Cs activity
Forest soil
Humus
Organic soil horizons
Clearcut
Shelterwood

ABSTRACT

The Chernobyl fallout of 1986 contaminated soils in Southern Germany. Levels of ^{137}Cs activity still remain high in forest soils. The contamination of forest food products still poses a serious problem for consumer health. Silvicultural interventions may offer an option to reduce the bioavailability of ^{137}Cs . The objective of this study is to investigate a possible effect of canopy opening and clearcutting as well as the selection of conifers and broadleaves for stand regeneration on the vertical soil profile of ^{137}Cs . The investigation uses a long-term silvicultural experiment on the artificial regeneration of a spruce stand 30 km southwest of the town Augsburg, Germany. 12 years after silvicultural intervention soil samples were taken horizon-wise and analysed to compare among the treatments. A suite of soil physical and chemical properties was determined per horizon including ^{137}Cs radioactivity, total carbon, texture, and cation exchange capacity. The results show that (i) the ^{137}Cs contamination at this site is high compared to other sites in Central Europe, (ii) regeneration cuttings did not markedly change the vertical ^{137}Cs profile in general within one decade, (iii) clear cutting modified the boundary between the organic soil layer and the mineral soil with expectable effects on the future ^{137}Cs bioavailability.

1. Introduction

The contamination potential of ^{137}Cs in forest ecosystems deriving from nuclear weapon tests as well as from accidents is still highly relevant. The Nuclear Power Plant (NPP) accident in Chernobyl happened some 30 years ago, a figure which also marks the half-life of ^{137}Cs . Beresford et al. (2016) reviewed lessons learnt in 30 years after the Chernobyl accident and five years after the Fukushima accident and dedicated a chapter to the ^{137}Cs behaviour in soils and sediments. High levels of ^{137}Cs originating from the NPP accident in Chernobyl were deposited on soils throughout Europe including Bavaria, Germany. The ^{137}Cs that entered the system with the Chernobyl fallout in 1986 was roughly 10-fold greater than ^{137}Cs deposition originating from nuclear weapon tests (Schimmack and Schultz, 2006).

Forest soils are particularly prone to high and long-term contamination by radionuclide materials compared to other land use systems. For example, a spruce stand has shown a 20% higher ^{137}Cs deposition than nearby grasslands (Bunzl et al., 1989). This is due to the high scavenging capacity of trees, especially that of dense canopy structures like coniferous forests compared to grasslands (Bunzl et al., 1989). Near Fukushima, soils under different land-uses displayed

similar ^{137}Cs levels, however the litter layer in forests presented higher values than herbaceous vegetation under other land-uses; moreover, ^{137}Cs penetrated deeper in forest soils (Koarashi et al., 2012a). The affected European forest soils are still highly contaminated even 30 years later, particularly in the organic and top soil layers (e.g. Winkelbauer et al., 2012b; Karadeniz et al., 2015; Pumpanen et al., 2016; Suchara et al., 2016). However, the fallout from nuclear weapons testing also contributes to this type of soil contamination. Highly bioavailable ^{137}Cs is retained in the organic soil layer (Karadeniz et al., 2015). The organic soil layer in forest ecosystems is characterized by a dynamic equilibrium between input in the form of litterfall and output as part of the decomposition processes. The properties of this layer greatly affect retention and bioavailability of ^{137}Cs , i.e. as organic matter has a high adsorption capacity due to the large surface area and the adsorption of ^{137}Cs cations on negatively charged compounds. A recent study in Japan also noted that organic matter appears to be the primary and preferential sorbent of ^{137}Cs (Teramage et al., 2016). ^{137}Cs is chemically analogue to potassium (e.g. White and Broadley, 2000) and its mobility in soil and availability to plants are positively correlated to organic matter content and cation exchange capacity of the soil, whereas it is negatively correlated to the clay content (Vanvoris et al.,

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1990). In this context, the impact of humus variability in terms of the layers themselves and their thickness becomes apparent (Völkel, 1994; Winkelbauer et al., 2011; Karadeniz et al., 2015). Thick humus layers can develop due to the complex interaction of poorly decomposable litter, low pH, poor potassium supply, and reduced microbial activity, (e.g. Kruyts and Delvaux, 2002; Winkelbauer et al., 2012b), however, climate is the main controlling factor. Humus layer thickness also plays a role in the observed reduced vertical migration into mineral horizons in forest soils (Rafferty et al., 2000; Winkelbauer et al., 2012b). Clay can effectively immobilize ^{137}Cs (Vanvoris et al., 1990; Bradbury and Baeyens, 2000) by storing it at the negative charged surface e.g. of illites and between the layers of swellable clay minerals like vermiculite and the smectite group. However, clay is mainly present in the underlying mineral horizons, and therefore ^{137}Cs must first pass the organic soil layer. In Japan, Nakanishi et al. (2014) showed that a large part of the ^{137}Cs moved from the litter layer into the mineral soil within a year, where it then would be controlled by factors such as pH, cation exchange capacity, organic carbon content, animal activity, soil texture, and type of clay minerals. However, particularly under K-depleted conditions, plant roots, which recycle ^{137}Cs and lead to the contamination of above-standing vegetation, and clay minerals act as competitive sinks (Kruyts and Delvaux, 2002). In spite of numerous studies, even decades after nuclear weapons testing and Chernobyl NPP accident the translocation mechanisms of ^{137}Cs in different types of forests are not fully understood (Pumpanen et al., 2016) and hence calls for further studies.

Since organic matter plays a very important role in the mobility of ^{137}Cs in forest ecosystems, changes in organic matter dynamics may strongly impact Cs cycling. Silvicultural interventions like clear cutting and reforestation are expected to induce changes in humus dynamics by reducing the input from litterfall and accelerating decomposition. Accordingly, they should manifest themselves in different ^{137}Cs migration trends depending on the type of intervention.

Many studies addressing forest management focus on the issue of contaminated timber, mushrooms, berries or game (e.g. Levula et al., 2000; Huotari et al., 2015). Others discuss options like tree-felling time, where early felling may prevent contamination of the core wood (Andersson and Roos, 2009); harrowing after thinning or clear cutting to improve tree growth and thus growth dilution (Shaw et al., 2001), or limiting tree harvest to areas with low contamination doses (Fesenko et al., 2005). Five years after clear cutting, Berthelsen et al. (1999) observed an increment of ^{137}Cs activity in organic soil layer, eluvial horizon and also its leaching from logging residues. Amiro et al. (1999) suggested to plough the top organic layer to physically relocate contamination to deeper soil layers thus decreasing the external surface dose and discussed that removing contaminated trees from the forest is an impractical countermeasure since new trees grown would still be contaminated. Hubbard et al. (2002) considered the options of thinning or clear-cutting along with deep ploughing or the physical removal of the organic layer to reduce contamination levels in forests. However, deep ploughing to bury contaminated material and remove it from the root zone of plants like mushrooms or berry bushes is an intervention that may simultaneously reduce soil fertility by removing the growth layer. Most importantly, ploughing is an option limited to only those forests, which can be accessed by the specific machinery (Hubbard et al., 2002). Völkel (1998) tested a variety of forest fertilizers to reduce ^{137}Cs uptake by plants in greenhouse pot experiments and found that calcium-rich fertilizers (Dolocorn, bentonites Calcigel and Montigel) were unsuitable, while potash-magnesia fertilizer (Patentkali®) stopped ^{137}Cs uptake by plants. Similarly, in field trials the trend was confirmed such that ^{137}Cs uptake reduced following potash-magnesia applications, while all other fertilizers were beneficial as well. Huotari et al. (2015) and Levula et al. (2000) reported a similar effect on the ^{137}Cs uptake of trees and shrubs by the application of wood ash. With renewed interest in the topic following the Fukushima NPP accident, it was stated that by removing aboveground tree biomass and all litter 80% of the

contaminated material in the study area could be decontaminated (Hashimoto et al., 2012), newly growing plants would continue to deliver contaminated litter though; hence, such actions presumably would need to be repeated multiple times. In fact, Teramage et al. (2014) observed that 45% of the ^{137}Cs deposited onto the forest floor during their study came from litterfall. However, the accelerated humus decomposition following interventions like clear-cutting (Berthelsen et al., 1999) does not remove contamination *per se* but allows for translocation. Guillitte and Willdrodt (1993) described that accelerated decomposition increased migration of radionuclides to mineral soil layers, where they would eventually be bound within clay minerals. They also indicated that ^{137}Cs mass activities reduced in fungal species in a recently cleared area compared to those growing nearby under forest canopy.

The main objective of this study is to examine the differences in the vertical distribution of ^{137}Cs in a forest soil in response to regeneration cuttings over time in order to assess silvicultural means reducing the bioavailability of ^{137}Cs . These treatments include clear cutting and shelterwood cutting and subsequent artificial regeneration with deciduous (*Fagus sylvatica* L.) and coniferous (*Abies alba* Mill.) trees. Investigations were carried out 12 years after the application of the treatments. Soil characteristics and ^{137}Cs mass and areal activity of the sampled plots were determined to identify the impact of the treatments. Additionally, ^{137}Cs mass activities of subsoil samples from the clearcut area from 2001 and 2013 were compared.

2. Material & methods

2.1. Study site

The study site is located 30 km southwest of the town Augsburg, Germany (48°08'N, 10°39'E). The average annual temperature and rainfall are 8.3 °C (meteorological station Landsberg am Lech, 1981–2010) and 900 to 950 mm (meteorological station Schwabmünchen, 1960–2010) (DWD, 2016). The evenly aged 93 years old stand is composed of 100% Norway spruce (*Picea abies* L.) and the research stand located within it has an area of 3.6 ha and lies 580 m a.s.l. on flat terrain (slope < 5%) (Felbermeier, 2004). The study site represents highly productive sites covered with spruce forest, which can be found on an area of about 170,000 ha in the Bavarian Tertiary Molasse Hills. The soil developed from a loess layer intermixed through solifluction and covers Miocene sediments from the Tertiary pre-alpine Molasse Basin (Felbermeier, 2004). Humus form is Moder; the soil was identified as Eutric Cambisol according to WRB/FAO (Zech et al., 2014).

Regeneration cuts were applied in December 2000: one block of the closed stand was cleared (CC) in the eastern part, one block was opened by shelterwood cutting (SC) by removing 15% of the basal area in the middle part, and one block was left without treatment (CN) in the western part. Each block has a size of around one ha and all above-ground biomass of the harvested trees was removed. In February 2001 the stand was group-wise (0.05 ha) replanted. Within each block three plots of beech (*Fagus sylvatica* L.), three plots of fir (*Abies alba* Mill.), and three control plots left for natural regeneration were established. In July 2001 three soil samples were taken from each plot (Felbermeier, 2004).

2.2. Sampling procedure

In July 2013 one plot with beech (P1–3) and one plot with fir (P4–6) – representing rejuvenation with deciduous and coniferous trees – were sampled in each block. The criteria for the plot selection are: (i) similar light conditions within the block (ii) no damages, and (iii) central location within each block. Additionally one control plot on the clearcut area, which was left to natural regeneration (P7), was also included in the sampling campaign. In 2013, the selected plots were fully covered

with beech regeneration (P1–P3), fir regeneration (P4–P6) and grass (P7).

Soil samples were collected from 5 locations within each plot and up to four horizons: the organic horizons Of, Oh and the mineral horizons Ah, Bv (German Soil Classification Symbols (Ad-hoc AG Boden, 2005)). For pits where the Ah horizon was comparatively thick, two samples of the Ah were collected (Winkelbauer et al., 2012b) and the mean of the measured values was used for the statistical analysis of this study.

Following the guidelines provided by Winkelbauer et al. (2012b) on each plot 5 soil pits (20 × 20 cm) were extracted with a spade to a depth of 30 cm and every horizon was sampled individually (sample unit). ¹³⁷Cs dynamics are intimately linked with humus decomposition properties and they differ for humus-rich and -poor horizons. Consequently, horizon-wise sampling was applied to provide the opportunity to relate findings to horizon-specific soil properties and compare between different locations with same horizons but varying thicknesses. The L horizons (litter) were excluded from sampling (c.f. Winkelbauer et al., 2012a). Taking into account previous experience significant differences can be expected at the humus-mineral soil interface, therefore gamma spectrometric and pedological analyses were performed on the four topmost samples.

A total of 103 samples were taken from 35 pits, with five pits extracted per plot. In some cases, very thin, irregular organic horizons < 3 mm could not be sampled, in other cases Of and/or Oh horizons were not present. For example, only in five sampling plots Oh horizons were present and sampled for radiologic measurements.

Additionally, archived soil samples collected in June 2001 were used to compare ¹³⁷Cs activity with that in 2013. The samples were taken at fixed depths from three locations within each plot (Felbermeier, 2004). In this study, we used the sampling depth of 5–10 cm taken from fir, beech and natural regeneration plots of the clearcut area corresponding to the Bv horizons of the sampling in the year 2013.

2.3. Laboratory procedure

Soil samples were dried to constant weight with air circulation chambers at a temperature of 40 °C. For the analysis of mineral horizons (Ah, Bv), the fine fraction (< 2 mm) was extracted by sieving, visible organic particles were removed. Organic horizons (Of, Oh) were shredded to a size < 1 cm with a customary grinder and well homogenized.

Pedological analyses included texture analysis following the combined pipette and sieving method of Köhn & Köttgen (clay: < 0.002 mm, silt: 0.002–0.063 mm, sand: 0.063–2 mm); pH was measured in 0.01 M CaCl₂; total carbon (C), total nitrogen (N), and total sulphur (S) contents were determined using an Elementar Vario EL III; effective cation exchange capacity (CEC) was measured with an atomic absorption spectrometer (Thermo Scientific™ iCE™ 3000 Series) after the sample was mixed with 1 M NH₄Cl dissolution. A carbonate test with 10% HCl was employed to determine the presence of carbonates. For a subset of the samples, CaCO₃ contents were derived with the gas volumetric Scheibler method (Calcimeter) and used to calculate the contents of inorganic carbon (IC). From total (TC) and inorganic carbon, the content of organic carbon (OC) can be derived mathematically (Prietz and Christophel, 2014).

For gamma spectrometric analysis, all samples, those taken in 2001 as well as those taken in 2013, were measured after the sampling in 2013. Most of the samples were measured in local facilities by the use of a Canberra Packard GC2519 high purity germanium detector (Table 1). Depending on available sample quantity 20, 50 or 100 ml of soil were measured in standard rubber bottles. Runtime was limited to either 1000 counts (often reached by samples from organic soil layers in under 24 h), assuring less than 5% uncertainty (1 Sigma) or a maximum runtime of 24 h (this runtime was often encountered for samples from greater depths). Data processing was administered by the software

package Genie™ 2000 (more detail on calibration and data processing can be found in Winkelbauer et al. (2012b)). To accelerate data acquisition, several samples were sent to the Radiation Protection Laboratory, Bayerisches Landesamt für Umwelt LfU (Bavarian Environment Agency), where samples were measured as well (Table 1). Similar configurations assure data comparability and results were double-checked by remeasuring some samples (with high, low and medium activities) with the local detector. Sample type and counting geometry affect minimum detectable activity (MDA) of measurements; ranging from 2.61 to 15.22 Bq kg⁻¹, mean MDA generally improves with increasing geometry and vertical depth (Winkelbauer et al., 2012b). ¹³⁷Cs areal activity densities were determined based on the recorded mass activity densities using sample weight and estimated bulk densities. These estimated bulk densities are 0.2 g cm⁻³ in Of, 0.4 g cm⁻³ in Oh, 1.0 g cm⁻³ in Ah, and 1.5 – SOM × 0.03 g cm⁻³ in mineral Bv horizons, respectively (for Of, Oh, Ah based on Umweltministerium Baden-Württemberg (1995), for Bv based on (Renger et al., 2008)). Areal activity density was then calculated by multiplying ¹³⁷Cs mass activity density, bulk density and horizon thickness.

2.4. Statistical analysis

Oneway ANOVA is applied for interference statistics using IBM SPSS Version 22. The model assumptions are proofed by Shapiro-Wilk's test for normal distribution and Levene's test for homoscedasticity. The Gabriel and Welsch Q and Hochberg's generalized Tuckey procedure were used for equal and unequal sample size, respectively. Brown-Forsythe statistics and Games-Howell's test are used in case of heteroscedasticity. In case of non-normality the Kruskal-Wallis procedure and Dunn-Bonferroni's test are applied (Field, 2009). Model equations are calculated by simple linear regression. Significance levels are defined as *p-value < 0.05, **p-value < 0.01, and ***p-value < 0.001.

3. Results

3.1. Basic soil properties

The mineral horizons, Ah and Bv, are all high in silt content with the lowest values found in P3, where a comparatively higher sand content was determined (Table 2); the soil textures fall into the categories of (clayey) loams. Across all treatments, soil pH values range from 2.91 in Ah horizons up to 5.27 in Bv horizons. The effective cation exchange capacity, CEC_{eff} (mmolc/100 g) decreases throughout the soil profiles, the averages in the individual horizons across all treatments are 20.6 in Of, 14.5 in Oh, 9.8 in Ah and 8.3 in Bv. CEC_{eff} is positively correlated with TC (Table 2), as can be seen in Fig. 1; moreover the aforementioned depth distribution becomes apparent.

The HCl-tests revealed isolated occurrences of carbonates in some samples in the form of single grains. However, due to the low CaCO₃ content determined on a subset of the samples, the mathematically derived values for organic carbon, OC, content were nearly identical to TC content. The average value determined for inorganic carbon, IC, is 0.11% (maximum is 0.23%). As some horizons were very thin, enough sample could not be collected, therefore certain analyses were prioritized. Since CaCO₃ was not expected in all samples according to the pre-test, this analysis was not done for all samples and TC is used in further discussions; especially in respect to the high TC contents in organic soil layers IC content is extremely small and, hence, negligible.

3.2. ¹³⁷Cs mass and areal activity densities of the research site

Measurements of mass activity densities are available for 103 samples from four different types of horizons in the organic soil layer (Of, N = 26 and Oh, N = 5) and mineral soil (Ah, N = 37 and Bv, N = 35).

To examine the depth distribution of ¹³⁷Cs in the different horizons,

Table 1

Technical equipment specifications. Given are the HPGe detectors used for gamma spectrometry indicating resolution and efficiency performances at 1.33 MeV.

Resolution (1.33 MeV)	Rel. efficiency (at 1.33 MeV) (%)	Detector model	Facility
1.9 keV	25	Canberra Packard, GC2519-7500	Technical University of Munich, GeoBoku
1.9 keV	40	EG + G Ortec, GMX-40195-S	Bavarian Environment Agency, Radiation Protection Laboratory, Southern Bavaria
2.2 keV	30	EG + G Ortec, GEM-30220-S	
1.9 keV	34	EG + G Ortec, GEM-33200	
2.0 keV	32	EG + G Ortec, GEM-33200	
1.8 keV	30	Canberra Packard, GC3018-7500	
1.8 keV	30	Canberra Packard, GC3018-7500	
1.8 keV	40	Eurisys Measures, EGPC 40-190-R	
1.8 keV	40	Canberra, GX4018	

measured values of all plots were used. The highest mass activity densities were determined in Oh horizons, followed by samples from Ah horizons, the overall lowest values were found in Bv horizons (Fig. 2a). However, when taking horizon thickness and estimated bulk density into account, the image shifts and in terms of areal activity density, Ah horizons feature the highest values, while those in Of, Oh and Bv are all comparatively low (Fig. 2b).

3.3. ¹³⁷Cs activity and regeneration cutting

Mean values of ¹³⁷Cs mass activity density of the Bv horizons increase from the control to the clearcut (Fig. 3a) with a significant difference between control versus shelterwood and control versus clearcut (Table 3). ¹³⁷Cs sample means of the Ah tend to display decreasing values from the control to the clearcut (Fig. 3), but without significant differences between the types of intervention. Mean ¹³⁷Cs values of the Of horizon have a maximum in the shelterwood cutting area (Fig. 3a), with a significant difference versus the control (Table 3). ¹³⁷Cs mass activity density is similar between plots planted with beech versus fir seedlings (Fig. 3a). Similar patterns and levels of significance were derived for ¹³⁷Cs areal activity (Fig. 3b, Table 3). Very high ¹³⁷Cs areal activity values are detected for the Ah horizon, which is the result of a combination of high ¹³⁷Cs mass activity, horizon thickness and bulk density of the horizon. On the clearcut, missing Oh layers and high ¹³⁷Cs levels in the Ah indicate intermingling of Oh material into the Ah horizon (Fig. 3).

The comparison of the ¹³⁷Cs mass activity density in the Bv horizon on the clearcut (including the natural regeneration) between the years 2001 ($\bar{X} = 61.6$ S.E. = 13.2 Bq/kg, n = 8) and 2013 ($\bar{X} = 94.3$ S.E. = 17.6 Bq/kg, n = 15) shows no significant increase of the mean.

Table 2

Basic soil properties ($\bar{X} \pm$ S.E.) of the different treatments (properties marked with * are average values for mineral soil, Ah & Bv).

Plot	Treat-ment	Sand* (%)	Silt* (%)	Clay* (%)	pH* (CaCl2)	CEC _{eff} * (mmolc/100 g)	TN* (%)	TC (%)			
								Of	Oh	Ah	Bv
P1	CC Beech	22.2 ± 1.2	51.2 ± 1.0	26.6 ± 1.2	3.6 ± 0.04	11.1 ± 0.6	0.36 ± 0.08	32.8 ± 5.4	-	9.8 ± 1.6	2.6 ± 0.4
P2	SC Beech	25.8 ± 2.0	49.4 ± 1.2	24.8 ± 1.1	3.4 ± 0.04	9.6 ± 0.5	0.50 ± 0.12	33.1 ± 4.1	-	15.8 ± 2.0	3.1 ± 0.3
P3	CN Beech	43.2 ± 2.0	33.6 ± 1.3	23.2 ± 0.9	3.4 ± 0.07	8.3 ± 0.3	0.27 ± 0.06	42.1 ± 3.0	36.0 ± 6.2	9.0 ± 1.5	2.2 ± 0.1
P4	CC Fir	24.4 ± 0.9	52.8 ± 0.4	22.8 ± 0.7	4.0 ± 0.06	7.5 ± 0.3	0.23 ± 0.04	-	-	5.7 ± 0.9	1.5 ± 0.2
P5	SC Fir	26.2 ± 1.0	48.8 ± 0.5	24.9 ± 0.6	3.4 ± 0.03	9.2 ± 0.7	0.35 ± 0.06	31.2 ± 2.1	16.2	9.6 ± 0.9	3.9 ± 1.0
P6	CN Fir	25.4 ± 0.6	49.6 ± 0.3	25.1 ± 0.7	3.3 ± 0.05	9.9 ± 0.4	0.30 ± 0.07	44.6 ± 1.2	27.7	9.3 ± 2.1	2.1 ± 0.2
P7	CC Natural	21.6 ± 0.7	52.2 ± 0.4	26.2 ± 0.8	3.7 ± 0.05	7.2 ± 1.0	0.37 ± 0.10	38.0 ± 2.7	-	10.6 ± 2.2	2.1 ± 0.8

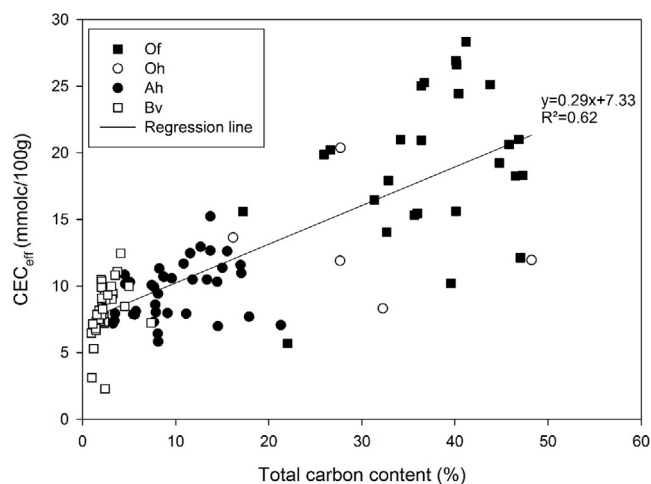


Fig. 1. Effective cation exchange capacity, CEC_{eff}, as a function of total carbon content for the different horizons: Of, Oh, Ah and Bv. The regression line is based on data from all horizons.

4. Discussion

The record of 103 gamma spectrometric measurements provides detailed information regarding the ¹³⁷Cs distribution throughout the forest soil profiles in the research stand. The forest stand belongs to the highly contaminated sites in Bavaria (e.g. compared to those described by Winkelbauer et al. (2012b)) with a mean ¹³⁷Cs activity of 27.4 kBq m⁻² and a range between pits of 6.7 and 66.8 kBq m⁻².

4.1. ¹³⁷Cs activity and regeneration cutting

Our observations document a significantly lower ¹³⁷Cs activity of

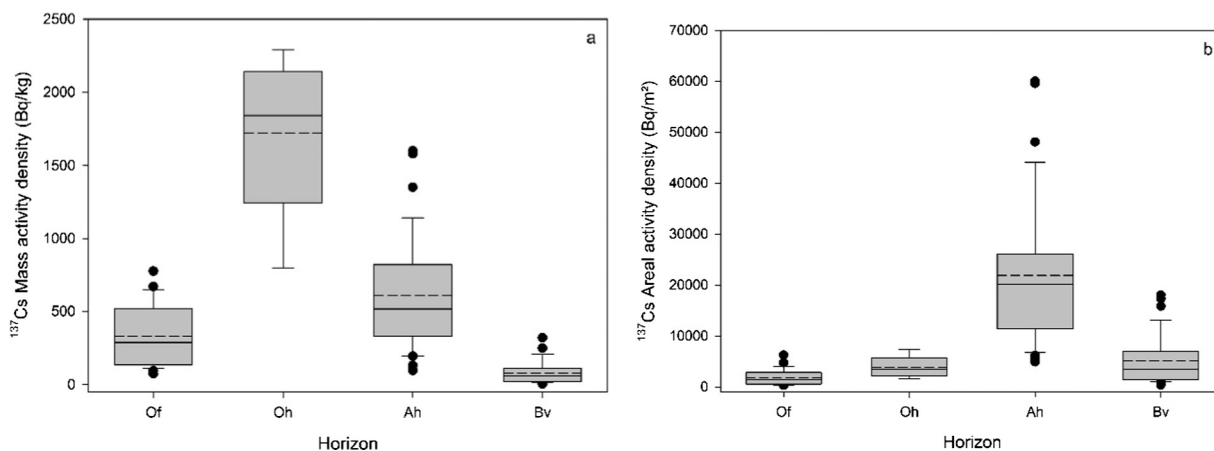


Fig. 2. Boxplots of ¹³⁷Cs of all plots in the different horizons in (a) mass activity densities (Bq/kg) and (b) areal activity densities (Bq/m²). Graph whiskers: 0.1 and 0.9 percentiles; box edges: 0.25 and 0.75 quartiles; solid box division: median (0.5), dashed box division: mean.

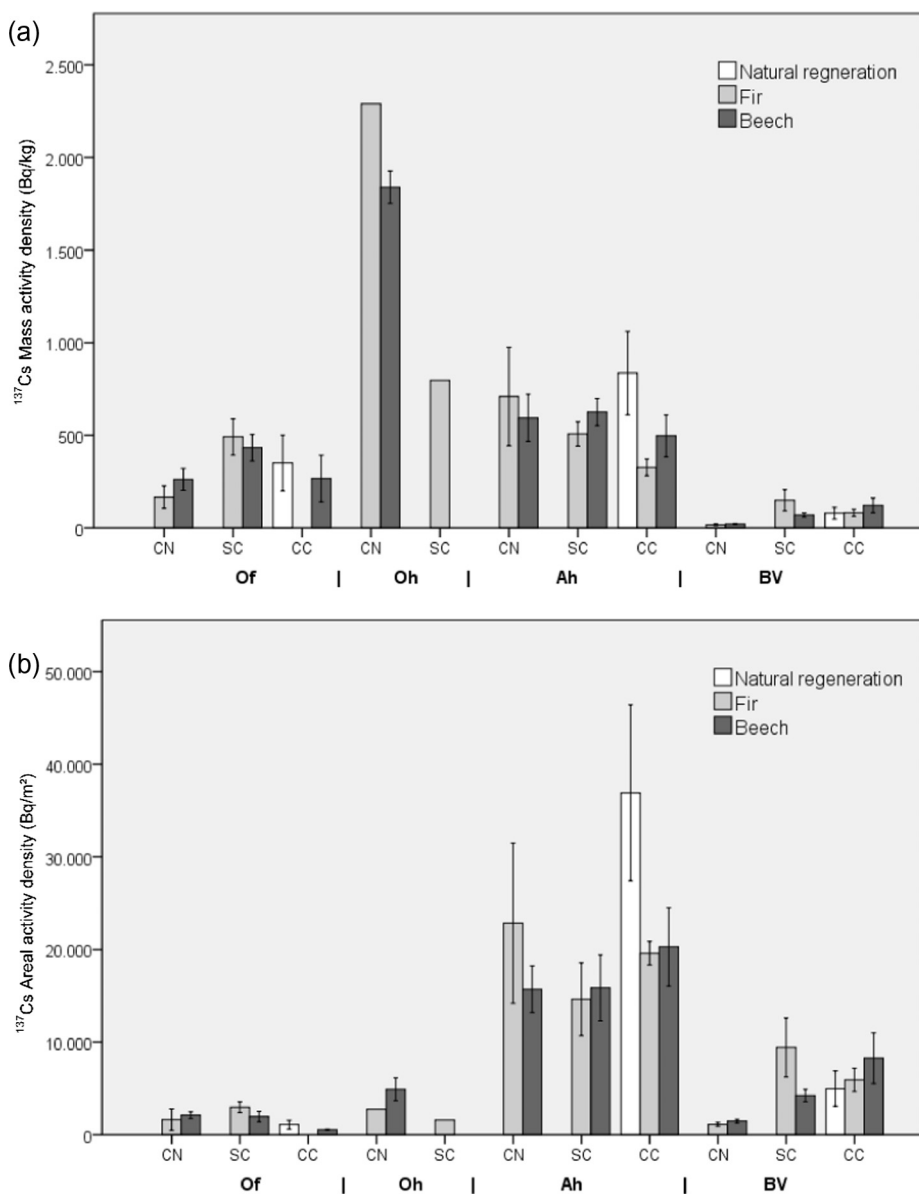


Fig. 3. Mean and S.E. of ¹³⁷Cs mass (a) and areal (b) activity densities of soil horizons for different silvicultural treatments (CN = control, SC = Shelterwood cut, CC = clearcut).

Table 3

Result of the ANOVA for ^{137}Cs activity in Of, Ah and Bv versus canopy opening (applied test statistics in brackets: ¹Fisher, ²Brown-Forsythe. Posthoc tests: ⁴Hochberg's generalized Tuckey, ⁵Games-Howell).

Horizon	Hypothesis	^{137}Cs mass activity	^{137}Cs areal activity
Of	$\mu_{\text{CC}} = \mu_{\text{SC}} = \mu_{\text{CN}}$	* (1)	* (1)
	$\mu_{\text{CC}} = \mu_{\text{SC}}$	n.s.(4)	n.s.(4)
	$\mu_{\text{SC}} = \mu_{\text{CN}}$	* (4)	n.s.(4)
	$\mu_{\text{CC}} = \mu_{\text{CN}}$	n.s.(4)	n.s. (4)
Ah	$\mu_{\text{CC}} = \mu_{\text{SC}} = \mu_{\text{CN}}$	* (2)	n.s.(2)
	$\mu_{\text{CC}} = \mu_{\text{SC}}$	n.s.(5)	n.s.(5)
	$\mu_{\text{SC}} = \mu_{\text{CN}}$	n.s.(5)	n.s.(5)
	$\mu_{\text{CC}} = \mu_{\text{CN}}$	n.s.(5)	n.s.(5)
Bv	$\mu_{\text{CC}} = \mu_{\text{SC}} = \mu_{\text{CN}}$	** (2)	** (2)
	$\mu_{\text{CC}} = \mu_{\text{SC}}$	n.s.(5)	n.s.(5)
	$\mu_{\text{SC}} = \mu_{\text{CN}}$	** (5)	** (5)
	$\mu_{\text{CC}} = \mu_{\text{CN}}$	* (5)	** (5)

the Bv-horizon in the control block, where no regeneration cuts were applied. Vertical translocation of topsoil ^{137}Cs into the Bv caused by forest canopy opening and removals must therefore be considered. A reason can be the mobilization of ^{137}Cs stored in the humus. It is known (e.g. Winkelbauer et al., 2012b) and also documented by our observations that high amounts of ^{137}Cs are stored in the humus. Humus itself responds sensitive to forest management. Physical intermingling of humus into the mineral soil by an increased faunal activity as well as the decay of soil organic matter (e.g. Likens, 2013) are processes that mobilize ^{137}Cs . Dissolved ^{137}Cs can be transported into the mineral soil until ^{137}Cs is fixed by the exchange surfaces of clay minerals (Suchara et al., 2016). The observed significant lower humus layers on the clearcut compared to the control block support the hypotheses that at least intensive regeneration cuts especially clear cutting influence the vertical distribution of ^{137}Cs . Following this hypothesis intensive regeneration cuts would have the potential to reduce bioavailability of ^{137}Cs .

Nevertheless, a comparison of 2001 versus 2013 shows no significant increase of ^{137}Cs activity in the Bv, which is contrary to this hypothesis. Therefore, the following aspects have to be taken into consideration before making any conclusions: (i) The Bv of the treated blocks versus the control block is characterized by a significantly higher clay content (Tables 4 and 5). This might improve the absorption of ^{137}Cs – especially for dissolved ^{137}Cs vertically transported in micro-channels. Consequently, the mineral characteristics can be one reason for the significant difference of ^{137}Cs mass activity of the control versus intervention. (ii) Cation concentration and base saturation of the Bv are significantly higher on the clearcut area. This can be an effect of the lower soil acidity (Table 2, 4 and 5) leading to a higher absorption of dissolved cations including ^{137}Cs in the Bv.

The results of this research and of other authors indicate that at least small canopy gaps and related silvicultural treatments have no large effect on the vertical ^{137}Cs distribution (e.g. Suchara et al., 2016). More intensive silvicultural interventions and humus as well as soil characteristics may have effects on the mobility and bioavailability of ^{137}Cs

Table 4

Mean and S.E. of humus layer thickness and Bv soil characteristics of the blocks ($\bar{X} \pm \text{S.E.}$, sample size available for statistical analyses in brackets).

	Control	Shelterwood	Clearcut
Humus layer thickness (cm)	4.2 \pm 0.7 (10)	3.0 \pm 1.2 (10)	0.7 \pm 0.9 (10)
pH	3.5 \pm 0.1 (10)	3.5 \pm 0.1 (10)	3.8 \pm 0.2 (10)
Base-cation saturation ratio (%)	6.4 \pm 0.3 (9)	13.1 \pm 1.5 (10)	17.4 \pm 1.1 (10)
Potassium (ppm)	2.9 \pm 0.1 (10)	4.7 \pm 1.2 (10)	7.4 \pm 0.6 (10)
Clay content (%)	23.2 \pm 0.7 (9)	26.2 \pm 0.4 (10)	26.7 \pm 1.0 (10)

Table 5

Result of the ANOVA for humus layer thickness and Bv soil characteristics versus canopy opening (applied test statistics in brackets: ¹Fisher, ²Brown-Forsythe, ³Kruskal-Wallis. Posthoc tests: ⁴Gabriel and Welsch Q, ⁵Games-Howell, ⁶Dunn-Bonferroni).

Hypothesis	Humus layer thickness (Sig.)	pH (Sig.)	Base-cation saturation ratio (Sig.)	Potassium (Sig.)	Clay content (Sig.)
$\mu_{\text{CC}} = \mu_{\text{SC}} = \mu_{\text{CN}}$	*** (3)	*** (2)	*** (2)	*** (1)	** (3)
$\mu_{\text{CC}} = \mu_{\text{SC}}$	*** (6)	*** (5)	n.s. (5)	*** (4)	n.s. (6)
$\mu_{\text{SC}} = \mu_{\text{CN}}$	n.s. (6)	n.s. (5)	*** (5)	*** (4)	** (6)
$\mu_{\text{CC}} = \mu_{\text{CN}}$	*** (6)	*** (5)	*** (5)	*** (4)	* (6)

at least on the long term.

4.2. ^{137}Cs bioavailability

^{137}Cs can be associated with clay and organic matter. Clay can effectively immobilize ^{137}Cs by storing it at the negative charged surface e.g. of illites and between the layers of swellable clay minerals (vermiculite and smectite). Organic matter instead possesses large surface area and therefore has a high ^{137}Cs adsorption capacity. According to Rigol et al. (2002) in organic soils (OM > 95%) with negligible clay contents the adsorption occurs mostly in non-specific-sites. They continue to outline that after a contamination event, two main factors account for the high transfer: the low solid-liquid distribution coefficient, which is due to the low clay content and high NH_4^+ concentration in the soil solution, and the low K^+ availability, which enhances root uptake (Rigol et al., 2002). In the context of bioavailability and mobility clay content therefore has a negative correlation while organic matter, and the associated cation exchange capacity, are positively correlated with ^{137}Cs . Moreover, since ^{137}Cs and potassium are analogues, plants may take up ^{137}Cs instead of potassium. In fact, plant roots compete with available selective sorption sites for ^{137}Cs , the root uptake leads to bio-recycling (Konopleva et al., 2009). Calmon et al. (2009) reviewed ^{137}Cs transfer parameter values in forest ecosystems and provided an overview of the transfer to trees, mushrooms, berries, game and reindeer. Pröhl et al. (2006) showed decreasing ^{137}Cs contaminations with ecological half-lives of about 12 a in game, forest plants and mushrooms for forests in middle Europe.

In the case of forest soils, ^{137}Cs , which was initially deposited on the surface, must first pass through the organic soil layer, before it can possibly be immobilized by clay, which is mainly present in the underlying mineral horizons. This study found high ^{137}Cs activities in the organic layer (Fig. 2), which in turn means that large amounts of ^{137}Cs are bioavailable. Also the Ah horizon contains high ^{137}Cs activities, however, in this horizon with larger clay content (than in the organic layer), more ^{137}Cs is likely immobilized. In the Bv horizon, organic matter contents are lowest and hence, compared to organic layer and Ah horizon, less ^{137}Cs can be adsorbed to it and remain bioavailable; instead a larger portion of the ^{137}Cs in this horizon presumably is fixed to clay minerals.

The low availability of clay minerals and the high content of organic matter turn the organic layer into a source of bioavailable ^{137}Cs . Thus, it becomes clear that apart from natural decay only vertical translocation (downward movement of organic matter and chemical exchange between organic and hydrous phase (Dorr and Munnich, 1989)) along with immobilization, or removal of contaminated biomass, can affect the plant-available pools of ^{137}Cs in the soil and especially the organic layer.

4.3. Organic soil layer properties

Organic soil layers play an important role for ^{137}Cs retention and availability in forest soils (Vanvoris et al., 1990; Völkel, 1994; Drissner

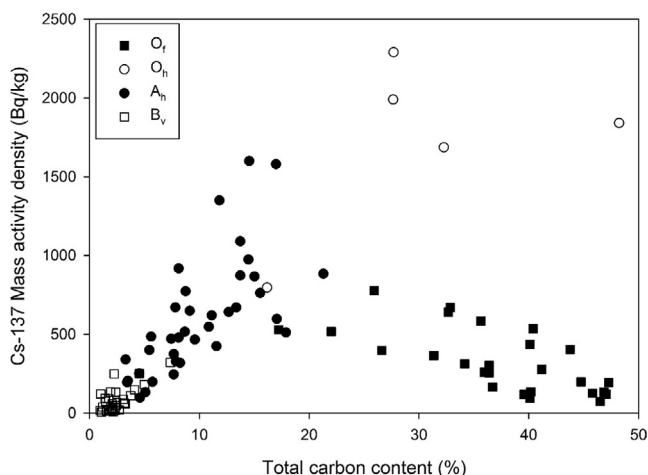


Fig. 4. ^{137}Cs mass activity density as a function of total carbon content of all plots for the different horizons: Of, Oh, Ah and Bv.

et al., 1998; Rafferty et al., 2000; Kruyts and Delvaux, 2002; Kruyts et al., 2004; Konopleva et al., 2009). From the different humus fractions, ^{137}Cs is particularly well associated with lignin (Andolina and Guillitte, 1990), a fraction that decays rather slowly. The organic soil layer consist of several horizons, differing in the degree of humification. The initially contaminated material at the surface decomposes over time and the adsorbed ^{137}Cs is released and may migrate to deeper horizons; however, other chemical processes may desorb ^{137}Cs as well. The different organic soil horizons (L, Of, Oh) have different decomposition rates. Depending on these rates they can thus retain ^{137}Cs in the organic soil layer for longer periods of time (Konopleva et al., 2009). Especially Oh horizons were found to effectively store ^{137}Cs very long (Winkelbauer et al., 2012b), as more humified horizons retain most ^{137}Cs (Karadeniz et al., 2015). For forest surface soils (0–3 cm) affected by the Fukushima NPP accident in Japan it was shown that ^{137}Cs uptake by microorganisms plays a subordinate role for retention of potentially mobile ^{137}Cs compared to ion-exchange adsorption on non-specific sites of abiotic components (Koarashi et al., 2012b).

The delayed translocation of ^{137}Cs can take several decades (Rafferty et al., 2000) and is reinforced by the uptake and renewed release of contaminated material through animals and plants at the surface. Particularly coniferous litter decomposes rather slowly and forms thick organic layers that retain more ^{137}Cs than those in deciduous or mixed forest soils (Scheglov, 1997). This is exacerbated by the location of roots within the soil profile: in coniferous forest soils they are mainly located in the Oh horizon, whereas in mixed forest soils roots are mainly in the mineral Ah horizon (Konopleva et al., 2009).

As already mentioned in the previous section, the low availability of clay minerals and the high content of organic matter turn the organic layer into a source of bioavailable ^{137}Cs . Fig. 4 illustrates this slow process of ^{137}Cs translocation throughout the organic soil layer and deeper soil profile. While a positive linear correlation between total carbon content and ^{137}Cs in mineral horizons and organic Oh horizons can be seen, this is not the case for Of horizons. In spite of high carbon contents less contaminated material is found in the Of horizons, this suggests that a large part of the contaminated material was translocated, which was adsorbed to organic matter that decomposed. This indicates that biological uptake may not return more contaminated material to the surface than is translocated downwards at the same time. However, also the quality of organic matter may play a role, i.e. the lignin content. In line with the findings by Winkelbauer et al. (2012b) also in this study the highest ^{137}Cs activities were found in Oh horizons, which are more humified. For a Japanese deciduous forest ^{137}Cs downward fluxes continuously decreased within the four years of observation, suggesting rapid leaching from the organic soil layer and

subsequent immobilisation by clay minerals in the upper mineral soil (Koarashi et al., 2016). Therefore, the soil investigated by them only presents a temporary source for ^{137}Cs biorecycling (Koarashi et al., 2016). For this study site, its history as a spruce stand may also be relevant. As discussed above, coniferous litter decomposes slower and many roots are located in the organic layer, hence ^{137}Cs may have been kept bioavailable in the organic layer for a longer time than it would have in a deciduous forest.

Regarding the linearity of the correlation in Fig. 4, a similar observation was made by Takenaka et al. (1998) on mineral soil samples, but since they had no considerable impact of Chernobyl fallout in their soils and therefore assumed an upper limit value, they discarded a linear regression as unreasonable. The lower values found here in Of horizons might be an expression of (i) the downward migration of ^{137}Cs with organic matter decomposition and (ii) low additional inputs via litter fall and bioturbation. Measurements of six litter samples collected in 2013–2014 from differently rejuvenated plots revealed an average ^{137}Cs mass activity density of 11.7 Bq/kg (median = 13.6 Bq/kg). This shows that trees continue to recycle ^{137}Cs , which they take up via their roots and return to the soil surface. Measured contamination levels were similar for litter samples from stands rejuvenated with beech or fir.

5. Conclusions

^{137}Cs contamination of the research stand is high compared to other forest sites in Bavaria. Following the nuclear weapons testing and 30 years after the Chernobyl NPP accident most of the ^{137}Cs is still bound to organic matter and therefore bioavailable – both for plants and game.

12 years after silvicultural intervention no general significant differences of the entire vertical soil profile of ^{137}Cs can be found for the investigated treatments. There exists no evidence that tree species selection for artificial regeneration - deciduous versus coniferous - has an influence on the vertical distribution of ^{137}Cs as well as ^{137}Cs activity of single horizons. However, the soil parameters suggest an increased ^{137}Cs translocation after intense opening or clearing of the forest canopy. It can be expected, that the modification of humus layers observed on the clearcut area will influence the ^{137}Cs mobility in the future.

The monitoring of the research site has to be continued with a higher sampling intensity to detect mechanisms of ^{137}Cs fixation in forest soils and for the development of silvicultural means to reduce the wide scale problem of ^{137}Cs bioavailability in the forest.

Acknowledgments

The authors thankfully acknowledge the support of Dr. Jennifer Winkelbauer (formerly TUM) managing the initial phase of the project.

We are grateful to the Radiation Protection Laboratory, Bayerisches Landesamt für Umwelt LfU (Bavarian Environment Agency), which measured several samples to accelerate data acquisition.

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