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Long-term migration of ^{137}Cs in Swedish grassland soil profiles following the Chornobyl accident

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ABSTRACT

This study presents long-term findings (1987–2008) on the vertical migration of ^{137}Cs from the Chornobyl accident in undisturbed grassland soils in central and northern Sweden. We examined five mineral and three organic soils, with ^{137}Cs deposition in 1986 ranging from 16 to 190 kBq m^2. ^{137}Cs activities were measured in 1 cm slices at depths of 0–10 cm and in 2.5 cm slices at 10–50 cm. Distribution (kBq m^2) was calculated for different soil horizons, and migration rates were determined based on observed depths. During the initial period after the fallout (1987–1992), ^{137}Cs was primarily located in the upper 0–2 cm layers of both mineral and organic soils, comprising 77 % to 94 % of the radionuclide. During the intermediate period (1994–2003), the average migration depth was 4.0 cm in mineral soils and 5.5 cm in the organic soils while during the later period (2004–2008) it was 4.4 and 7.0 cm, respectively. After about 20 years, approximately 80 % of the ^{137}Cs activity was found in the upper 0–6 cm at five out of eight sites, and 75–78 % within 0–9 cm at two sites regardless of soil type. The average radionuclide migration rate of ^{137}Cs in the period 1987 to 2008 across various sites was 0.31 cm yr$-^1$. In mineral soils, the average migration rate was 0.28 cm yr$-^1$ (range 0.13–0.47) and in organic soils it was 0.3 cm yr$-^1$ (range 0.17–0.76). There was no significant relationship between soil clay content in mineral soils and the average migration depth of the radionuclide during the study period ($P = 0.423$).

1. Introduction

The knowledge of ^{137}Cs migration within soil profiles from the long-term perspective is important for prediction of radionuclide variation and accumulation in soil and plants. Such accumulation and distribution will affect the ambient dose rate as well as potential radiation exposure to man and requires planning of appropriate countermeasures aiming to reduce the root uptake of radionuclide by grassland species and agricultural crops, and thus the transfer to the humans through the food-chain.

As a result of the accident at the Chornobyl nuclear power plant on April 26, 1986, large areas in the eastern coastal areas of central and northern Sweden were heavily contaminated with radionuclides. Amongst other radionuclides radiocaesium (^{137}Cs) was deposited in concentrations of more than 100 kBq m^{-2} (Andersson et al., 2007).

Following the fallout on the vegetation and the surface of grassland soils, the radionuclide initially accumulates in the litter layer or at the soil surface, leading to high levels of radioactivity that decline in subsequent years (Varskog et al., 1994). Over time, radioactive caesium began migrating into the upper soil layers, which represent the primary rooting zone for grassland plant species, allowing it to be absorbed by plants through their root systems (Vinichuk et al., 2025). The migration rate of ^{137}Cs within the undisturbed grassland soil profile depends mainly on the soil properties and is generally fastest in sandy loam (Forsberg et al., 2000) and on the plant uptake and circulation of Cs in the topsoil-plant system (as a substitute of K), in particularly in nutrient poor soils (Rosén et al., 1999). Generally, ^{137}Cs is strongly adsorbed by 2:1 clay mineral (Delvaux et al., 2000) and a close relationship between soil fine fractions and ^{137}Cs mobility in soil profile has been reported (Bonazzola et al., 1993).

Eight years after the fallout, substantial amounts of ^{137}Cs had migrated to about 50 cm deep in moist organic and podzolic soils in heavily polluted regions of Ukraine, Belarus, and Russia (Ivanov et al., 1997). Seven years after the accident the majority (78–99 %) of the deposited ^{137}Cs in Belarus was in the top 10 cm of the soil (Arapis et al., 1997) and migration rates increased with depth (Knatko et al., 1996).

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The migration rate of ^{90}Sr within the soil profile has been shown to be higher than that of ^{137}Cs in sandy loam and loam soils (Forsberg et al., 2000).

The soil organic matter (SOM) content can have different effects on ^{137}Cs migration in the soil. Soils with high SOM often show high ^{137}Cs mobility within the soil profile (Sanchez et al. 1999) since organic compounds have only an indirect effect on ^{137}Cs binding, and adsorption occur mostly on non-specific sites (Rigol et al., 2002). Dissolved SOM bound to the soil adsorbs exchangeable ^{137}Cs, thereby reducing its migration within the soil, while dissolved SOM in the soil solution can release adsorbed ^{137}Cs, facilitating its movement through the soil profile (Tatsuno et al., 2020).

The migration rates of ^{137}Cs in undisturbed mineral and organic soil profiles are highly variable depending on soil type, land uses (Takahashi et al., 2015), management practices (Li et al., 2022), geographical locations but generally do not exceed 1.0 cm yr^{-1} during the first year after deposition (Rosén et al., 1999; Jagercikova et al., 2015). Other processes that influence the radionuclide movement within soil profile includes the pre-diffusion time of establishing physicochemical equilibrium in the system, soil moisture, concentration and composition of the soil solution and pH (Gosman & Blažiček 1994), as well as bioturbation (Jarvis et al., 2010).

The longer ^{137}Cs will remain in the upper soil horizons, the longer time it will be readily available for root uptake and transfer to vegetation. Due to root uptake of ^{137}Cs by plants radionuclide will remain in the upper soil horizons for longer time than in a bare soil.

There were several studies (Haak et al., 1973; Bachhuber et al., 1982; Wilkens et al., 1984) focusing on the distribution and migration of ^{137}Cs in undisturbed soils carried out in the pre-Chornobyl period. Even more research in both semi-natural and natural grassland areas have been done in the post-Chornobyl period (Isaksson & Erlandsson, 1995; Forsberg et al., 2000; Takahashi et al., 2015).

Meanwhile, studies of long-term vertical migration of ^{137}Cs in undisturbed grassland soils are rare and only a few are available (e.g. Kaissas et al., 2023). Data from relatively long-term studies (1987–1995) of Chornobyl originated ^{137}Cs migration in Swedish undisturbed soil profiles (Rosén et al., 1999) indicate that downward migration was most pronounced in the organic soils and in the podzols. In southern Sweden, over a 10-year period from 1988 to 1998, ^{137}Cs initially migrated through the topsoil to a depth of 2.8 cm in 1988, after which its migration slowed significantly (Isaksson et al., 2001). Studies on ^{137}Cs migration in 14 soil groups between 1992 and 2007 (Jagercikova et al., 2015) demonstrated that radionuclide penetration velocities ranged from 0.05 to 0.76 cm yr^{-1} (median of 0.28 cm yr^{-1}).

This study presents a follow-up to the previously published work (Rosén et al., 1999) on the long-term migration of ^{137}Cs in undisturbed Swedish grassland soil profiles (1987–1995).

Data for the period 1987–1994 (including the Stora Blåsjön site for 1995) are taken from the earlier publication. New data for the period 1995–2008 are presented here where also an additional site, Åsenmossen, has been added.

The aim of this study was to present and discuss the data on ^{137}Cs migration within eight grassland soil profiles during twenty (1987–2008) years. These results are compared and discussed as related to soil type and texture. We hypothesized that ^{137}Cs migration rate within soil profiles in a long-term perspective will decrease with time independent of soil types.

2. Materials and methods

2.1. Study areas

Eight experimental fields with typical soil types and soil texture classes were selected in corresponding areas with high deposition fall out levels of ^{137}Cs as a result of Chornobyl accident in 1986 (Fig. 1).

The selected fields were under temporary or permanent grasslands

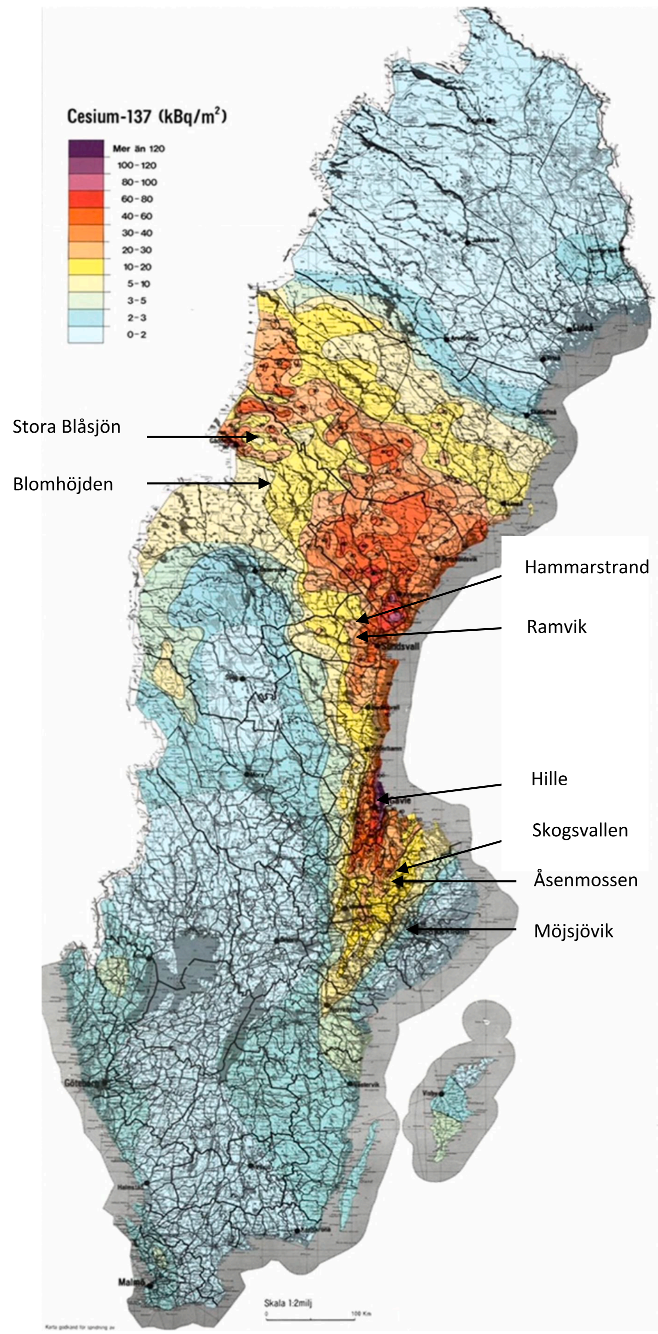


Fig. 1. Location of the eight Swedish study sites and the deposition of ^{137}Cs (kBq m^{2}) from the Chornobyl accident, based on inflight measurements performed from May to October 1986 (SGAB, 1986).

occasionally fertilised and used for hay production and semi-natural or natural pastures used for extensive grazing. The soils in the study sites have not been ploughed since 1986. The distribution of ^{137}Cs within soil profiles in those areas was studied in the period between 1996 and 2007/2008 by sampling and analysing soil samples from different layers to depths of 50 cm.

2.1.1. Site and soil descriptions

^{137}Cs migration was studied at the eight sites comprising different soil types according to FAO (1988) and Soil Survey Staff (1992), including five mineral soils and three organic soils (Table 1, Supplementary Material Table S1). Table S1 also presents 30-year average (1991–2020) annual precipitation and mean temperature data from the

Table 1
Selected properties of the studied soils profiles (modified from Rosén et al. 1999).

#	Horizon	Depth, cm	pH_{H2O}	Bulk density, $g\ cm^3$	Particle size distribution (mm), % sand 2–0.05	silt 0.05–0.002	clay < 0.002	C, %	N, %	K-AL, mg 100 g^{-1}	K-HCL, mg 100 g^{-1}
1	Skogsvallen, Typic Dystrochrept (Soil Survey Staff, 1992); Dystric Cambisol (FAO, 1988)										
	Ah1	0–3	5.2	0.72	15	43	42	12.70	1.04	3.63	317
	Ah2	3–5	5.3	1.40	–	–	–	3.40	0.33	13.0	363
	Bw1	5–20	4.7	1.32	15	42	42	1.90	0.18	9.70	398
	Bw2	20–35	6.2	1.37	13	40	46	1.30	0.13	12.9	467
	BC1	35–50	6.5	1.39	12	40	48	1.30	0.13	13.3	460
	BC2	50–75	7.4	1.35	6	42	52	0.70	0.09	17.1	606
	C	75–95	7.7	–	1	45	54	0.20	0.04	17.6	637
	∑ (mean)		6.1	1.3	10.3	42.0	47.3	3.1	0.3	12.5	464
2	Möjsjövik, Typic Sulfihemist (Soil Survey Staff, 1992); Thionic Histosol (FAO, 1988)										
	Hap[a]	0–20	5.5	0.33	–	–	–	37.3	2.29	45.4	87
	He1	20–50	5.4	0.15	–	–	–	37.4	1.9	6.2	34
	He2	50–75	5.3	0.12	–	–	–	40.9	1.93	9.5	52
	Hi	75–85	5.3	0.10	–	–	–	42.5	1.79	5.0	64
	Har	85–100	5.0	0.14	–	–	–	28.6	2.08	7.5	149
	2Cr	100–110	5.4	0.39	–	–	–	–	–	14.9	402
	∑ (mean)		5.3	0.2				37.3	2.0	14.8	131
3	Hille, Terric Sulfihemist (Soil Survey Staff, 1992); Thionic (Terric) Histosol (FAO, 1988)										
	Hap[a]	0–25	5.9	0.34	–	–	–	26.7	1.74	6.8	36
	He	25–33	5.5	0.16	–	–	–	36.9	1.88	9	28
	Hi	33–47	4.3	0.19	–	–	–	44.7	2.07	8.2	15
	2Cg	47–55	3.1	0.29	–	–	–	11.5	1.2	7.4	190
	2Cr(g)	55–100	2.9	0.44	–	–	–	5.5	0.65	26.3	355
	∑ (mean)		4.3	0.3				25.1	1.5	11.5	124
4	Ramvik, Typic Udorthent (Soil Survey Staff, 1992); Eutric Regosol (FAO, 1988)										
	Ah1	0–7	5.4	–	47	42	12	4.2	0.3	27.2	145
	Ah2(Ap)	7–23	5.6	1.27	44	48	8	1.8	0.15	14.6	130
	Bw	23–34	5.6	1.41	32	57	11	0.8	0.07	6.6	165
	CBg	34–55	5.8	1.38	52	40	8	0.3	0.03	5.5	164
	Cg1	55–75	5.8	1.55	21	66	13	0.2	0.02	9.5	231
	Cg2	75–100	5.6	1.56	16	71	13	0.2	0.02	9.4	228
	∑ (mean)		5.6	1.4	35.3	54.0	10.8	1.3	0.1	12.1	177
5	Hammarstrand, Typic Udorthent (Soil Survey Staff, 1992); Eutric Regosol (FAO, 1988)										
	Ah1	0–6	6.3	0.68	34	56	10	4.13	0.29	51.4	102
	Ah2(Ap)	6–20	6.1	1.12	38	57	5	1.33	0.11	3.6	70
	Ah3	20–30	6.2	1.24	47	51	2	0.53	0.03	2.2	60
	C1	30–50	6.4	1.34	47	51	2	0.25	0.01	2.2	46
	C2	50–100	6.6	1.27	49	49	2	0.21	0.01	2.2	44
	∑ (mean)		6.3	1.1	43.0	52.8	4.2	1.3	0.1	12.3	64
6	Stora Blåsjön, Typic Cryorthent (Soil Survey Staff, 1992); Dystric Regosol (FAO, 1988)										
	Oi	0–2	5.9	0.91	–	–	–	16.6	0.97	69.8	103
	Ah	2–4	6.2	1.29	54	36	10	4.33	0.32	9.4	52
	Ah/E/B(Ap)	4–15	6.5	1.12	56	39	5	1.62	0.12	2.6	51
	Bs(Bw)	15–35	6.7	1.32	58	39	3	1.05	0.08	1.6	66
	Bc	35–60	6.0	1.55	58	39	3	0.56	0	1.4	69
	Cg	60–100	6.1	–	63	36	1	0.13	0	2.2	96
	∑ (mean)		6.2	1.2	57.8	37.8	4.4	4.0	0.2	14.5	72
7	Blomhöjden, Typic Haplocryod (Soil Survey Staff, 1992; not identified (FAO, 1988)										
	Oi	0–2.5	5.2	–	–	–	–	31.79	2.33	83.7	105
	Oe	2.5–5	4.5	0.44	–	–	–	24.46	1.63	42.4	60
	E	5–10	4.9	1.33	63	29	8	1.72	0.06	2.6	29
	Bs1	10–17	5.4	0.97	54	28	18	3.84	0.19	1.8	75
	Bs2	17–30	5.8	1.24	63	25	12	1.92	0.09	1.4	141
	C	30–70	6.5	1.49	60	31	9	0.36	0.02	2.2	272
	∑ (mean)		5.4	1.1	60.0	28.3	11.8	10.7	0.7	22.4	113
8	Åsenmossen, Fibric Histosol (Soil Survey Staff, 1992), not identified (FAO, 1988)										
	Hi1[a]	0–15	3.9	0.25	–	–	–	47.9	0.85	117.0	132
	Hi2	15–25	3.7	0.51	–	–	–	45.6	1.05	33.2	39
	He1	25–35	3.8	0.38	–	–	–	45.9	1.03	8.4	11
	He2	35–45	3.7	0.41	–	–	–	45.6	0.89	6.4	9
	He3	45–55	4.0	0.59	–	–	–	49.5	1.62	2.9	4
	∑ (mean)		3.8	0.40				46.9	1.1	33.6	38.9

[a] a = sapric (highly decomposed organic material); e = hemic (moderately decomposed organic material); i = fibric (slightly decomposed organic material); p = ploughing or other human disturbance; r = strong reduction. The horizon suffixes are followingFAO (2006).

nearest weather station (SMHI, 2025).

Skogsvallen is a permanent pasture on a silty clay soil classified as a Typic Dystrochrept (Soil Survey Staff, 1992) or Dystric Cambisol (FAO, 1988) and located about 50 km northwest of Uppsala (Västmanlands County). The sampled field is in a narrow valley between an esker and a moraine hill. The field has not been cultivated or fertilised since the 1960 s and the vegetation was presented by various grass species. The mean deposition at the site (sampled 1987–2007) was estimated as 91 kBq m^{-2}.

Möjsjövik is pastureland on a fen-peat soil classified as a Typic Sulfihemist (Soil Survey Staff, 1992) or Thionic Histosol (FAO, 1988) and located about 25 km west of Uppsala (Uppsala County). The organic (histic) horizons are 85 cm deep and overlay a sulphide rich mud. The mean deposition at the site (sampled 1987–2007) was estimated as 65 kBq m^{-2}. The sampled field has been used for pasture, and the vegetation consists of herbs and broad leaf grasses. The field has been ploughed but only prior to the Chornobyl fallout. In 1987 and 1988 the soil was fertilised with 50 kg ha^{-1} of potassium.

Hille is a temporary grassland on a fen-peat soil located about 10 km north of Gävle (Gävleborg County). The soil consists of 50 cm thick organic horizons, formed from fen-peat. Below the peat lies a layer of sulphide-rich mud. The mean deposition was estimated (sampled 1987–2007) as 190 kBq m^{-2}. The field is drained and was used for crop production before the Chornobyl accident and was at those times ploughed and fertilised regularity. The soil has not been cultivated since the fallout and the vegetation consists of different species of grass and small shrubs. The soil was classified as a Terric Sulfihemist (Soil Survey Staff, 1992) or Thionic (Terric) Histosol (FAO, 1988).

Ramvik is a permanent grassland on silty and sandy loam soil, located about 23 km northwest of Härnösand in Västerbotten County. The sampled field is located on a terrace in the upper part of a steep slope (30 %) with a mean deposition (sampled 1987–2007) of 53 kBq m^{-2}. The field was used as permanent pasture, and the vegetation consists of various grass species and herbs. The land has neither been ploughed nor fertilized since the late 1970 s. The soil is well drained, and roots are found down to 60 cm depth. The soil was classified as a Typic Udorthent (Soil Survey Staff, 1992) or Eutric Regosol (FAO, 1988).

Hammarstrand is a permanent grassland on a silty loam soil situated about 80 km east of Östersund (County of Jämtland). The sampled field is located on a gentle slope just by the Indals River and are subject to runoff from higher areas in some years. The mean deposition at the site was estimated (sampled 1989–2007) as 40 kBq m^{-2}. The soil has neither been ploughed nor fertilized since the middle of 1970 s and are currently used for pasture. The vegetation consists of various grass species and herbs. The soil was classified as a Typic Udorthent (Soil Survey Staff, 1992) or Eutric Regosol (FAO, 1988).

Stora Blåsjön is semi-natural grassland on a gravelly sandy loam soil located about 200 km northeast of Östersund (County of Jämtland). The area is mountainous, and the sampled field is situated on a steep slope at the base of Mesklumpen (924 m). The mean deposition at the site was estimated (sampled 1989–2007) as 34 kBq m^{-2}. The land has been ploughed on a few occasions during the 1950 s but was then used as permanent pasture. Various spices of grass and herbs grow on the site. The soil was classified as a Typic Cryorthent (Soil Survey Staff, 1992) or Dystric Regosol (FAO, 1988).

Blomhöjden is a mountainous area located about 150 km northwest of Östersund in the Jämtland County. It is a semi-natural grassland, which is used as extensive mountain pasture for cattle and goats. The soil is a podsolated gravelly sandy loam. The field is located on a gently sloping moraine hill. The mean deposition at the site was estimated to (sampled 1990–2007) 16 kBq m^{-2}. The vegetation consists of various types of grass and herbs. The soil was identified as Typic Haplocryod according to Soil Survey Staff (1992).

Åsenmossen is located about 40 km northwest of Uppsala in Uppsala County. The field is located on a former bog, which has collapsed due to excavation in the 1930 s. After the trenching, pine trees were planted on the entire area. Currently, most of the ditches are overgrown, except for a furrow in the middle where a lot of water flows in the spring. The mean deposition at the site (sampled 2000–2008) was 31 kBq m^{-2}. The undergrowth on the bog is dominated by a low to medium-sized evergreen shrub marsh Labrador tea (*Rhododendron* spp.) and various species of sphagnum mosses. The soil consists of sphagnum peat and decayed deciduous tree remains. The soil was classified as Fibric Histosol according to Soil Survey Staff (1992).

The first sampling was carried out between the end of August and beginning of November 1987 (Skogsvallen, Möjsjövik, Hille, Ramvik) followed by sampling in spring 1989 (Hammarstrand, Stora Blåsjön), in summer 1990 (Blomhöjden) and autumn 2000 (Åsenmossen).

2.2. Sampling techniques

2.2.1. Soil profiles

Soil profiles at each of the sampling sites were excavated to a depth of 110 cm. The description of the profiles was made according to the guidelines of the FAO (1990), and the classification of the soil according to the Soil Map of the World Legend (FAO, 1988) and Soil Taxonomy (Soil Survey Staff, 1992). Samples were taken from each horizon for soil chemical and physical analyses. To determine the bulk density, representative soil samples weighing approximately one kg and two cylindrical samples (55 cm^3) were taken from each soil horizon.

2.2.2. Soil cores

The soil cores were taken with 2–3 year's interval to a depth of 15–25 cm in 1987–1995 and up to 60 cm in 1998–2008. Soil cores were taken at three (in a few cases two or four) sub-sites (A, B, C) on a circle area of 78 m^2 with an interval of 20 m between in the longitudinal direction of the field (Fig. 2a). Five soil cores were collected from each sub-site in two stages. Firstly, using an auger (diameter 57 mm), soil cores were taken to a depth of 10 cm and sliced into 1-cm layers. Secondly, soil cores (diameter 22 mm) were taken from a depth of 10–50 cm and cut into 2.5 or 5.0 cm layers (Fig. 2b).

Soil samples from the same soil layer at each sub-site were pooled to form bulk samples. Sub-sites were used as replicates (2–4) for the statistical evaluation of results. In the laboratory, soil samples were air-dried (at a maximum of 40 °C) for one week and sieved through a 2 mm sieve before radiometric and soil analyses.

2.3. Methods of analyses

2.3.1. Soil analyses

Particle size distribution in mineral soils was analysed by sedimentation using the hydrometer and/or pipette method (Day, 1965). Soil particles sized 2–0.05 mm were classified as sand, 0.05–0.002 mm as silt, and those smaller than 0.002 mm as clay (Table 1). To determine the bulk density of the soil, duplicate cylinders were dried at 105 °C. Total C was analysed in duplicate on an elemental analyser by Leco instrument.

2.3.2. ^{137}Cs determination

^{137}Cs activity concentrations in aliquots of soil samples were determined using high-purity germanium detector systems located in a low-background laboratory. To obtain acceptably low counting errors for ^{137}Cs (1–5%), the measurement time varied from 1 to 48 h. The concentration of ^{137}Cs activity in soil samples was measured on a dry weight (d.w.) basis. Activity data were adjusted for decay to the April 26, 1986 (Chornobyl accident).

2.3.3. Calculations of ground deposition, migration rates and depth

^{137}Cs deposition in the soil profile was calculated using soil activity concentration measurements and soil bulk density determined at specific horizons. ^{137}Cs deposition at each horizon was calculated by dividing the total radionuclide activity in each layer by the surface area of the core and thereafter summed up.

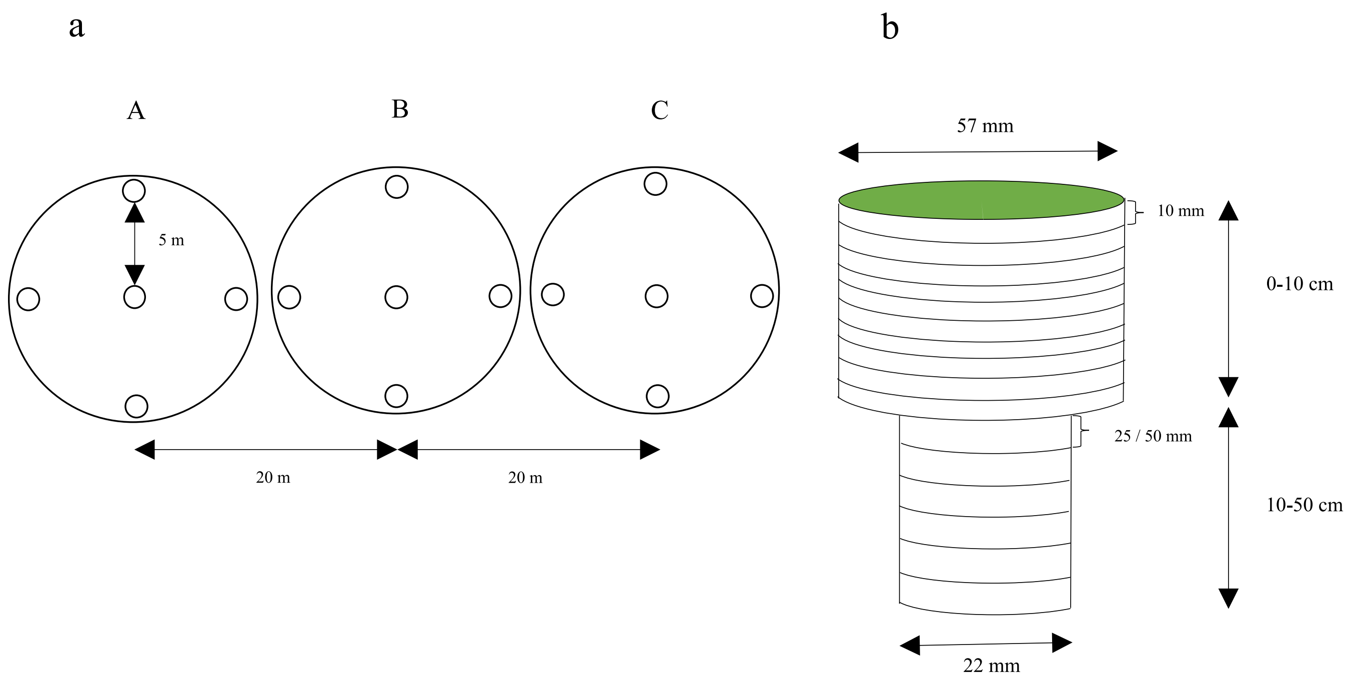


Fig. 2. Sampling of soil cores up to 50 cm depth at three sub-sites (A–C), with five bore holes in each sub-site, (circle area 78 m^2), a. The soil cores were sliced in 10 mm, 25 mm and 50 mm layers, respectively, b.

The migration depth was calculated for different sampling occasions. The migration rate of ^{137}Cs within the soil profile for the first year was estimated from the results of the first years of sampling. The average migration rates for subsequent years were calculated as the difference in average depth between the first and second, or first and third, sampling events, and then recalculated per unit of time (cm yr^{-1}). To compare the vertical migration of ^{137}Cs between different soils, independent of ground deposition, the relative activity (q_i) was calculated. The relative activity in each layer is described as a fraction between the activity in specific soil layer A_i (Bq m^{-2}) and the total activity at the site A_{tot} (Bq m^{-2}) (Eq (1):

$$q_i = \frac{A_i}{A_{tot}} \tag{1}$$

The migration can be described by calculating the migration depth (X), i.e. a weighted mean value for the activity in the soil, as described in (Arapis et al., 1997) (Eq (2).

$$\sum_{i=1}^{n} (X - X_i)q_i = 0 \tag{2}$$

where X_i represents the centre of each layer and q_i the relative activity of that layer. The migration rate is then calculated by dividing the change in migration depth (cm) with time (yr).

The average depth of the radionuclide was calculated as the sum of the relative deposition of the radionuclide in subsequent soil layers at different depths (Bq $m-^{2}$ / cm) per layer (X_i * q_i), divided by the sum of radionuclide quota of the total quantity corresponding to each soil layer (q_i).

The difference between the migration depths (X) ^{137}Cs in soil profiles was tested by using the non-parametric Mann–Whitney U test at $P < 0.05$. The statistical analysis was performed by using Minitab Statistical Software Version 2023 Minitab® 21.4.

3. Results

3.1. Soil properties

^{137}Cs migration and distribution were estimated in soil profiles of five mineral and three organic soils of different origin during the period of 1987–2008. Selected soil chemical and physical properties of the studied soils are presented in Table 1. The particle size distribution was determined to 110 cm depth (where possible) and varied widely. The silty clay soil at Skogsvallen consisted on average of 10 % sand, 42 % silt and 47 % clay. Particles size distribution in the loam and silty loam soils at Ramvik and Hammarstrand was rather similar with 35–43 % sand, 53–54 % silt and 4–11 % clay. The sandy loam soils at Stora Blåsjön and Blomhöjden had relatively high content of sand (58–60 %), moderate silt (28–38 %) and low clay (4–12 %) content. Based on the soil properties, the studied soils can be divided in two major soil type; (i) Mineral soils – Skogsvallen, Ramvik, Hammarstrand, Stora Blåsjön and Blomhöjden, and (ii) Organic soils – Möjsjövik, Hille and Åsenmossen with an average organic carbon content of 37, 25 and 47 %, respectively.

Exchangeable potassium (K-AL) within a depth of 20–25 cm ranged from 6.8 to 75.0 mg per 100 g of soil, with relatively low levels (6.8 to 8.3 mg) at the Skogsvallen and Hille sites and peaking at 75.0 mg per 100 g at the Åsenmossen site. In the mineral soils, the organic carbon content in the topsoil varied from 32 % in the moor layer of the podzol (0–7 cm) to about 4 % in the Regosols, and in the sub-soil horizons it did not exceed 1 %, with some exceptions in the spodic (Bs) horizons (Table 1). ^{137}Cs ground deposition in 1986 was ranging from 16 to 190 kBq m^{-2} (Table 2, Supplementary Material Table S2).

3.2. ^{137}Cs distribution, migration rate and depth in soil profiles over time

Radionuclide distribution within soil profiles was estimated as % of Bq/m^2/cm and determined first in August (Skogsvallen), September (Möjsjövik, Ramvik) and November (Hille) 1987, a bit more than one year after fallout. ^{137}Cs migration in soil profiles was compared across three periods: early (1987–1992), intermediate (1994–2003), and late (2004–2007).

The early period from 1987 to 1992. In 1987 (Figs. 3.1–3.8, Table 2, Supplementary Material S2), ^{137}Cs was primarily concentrated in the upper soil layers, with 77 % (Hille) to 94 % (Skogsvallen) of the radionuclide found at depths of 0–2 cm in both mineral and organic soils. Migration depth ranged from 0.89 cm at Skogsvallen to 2.22 cm at Hille, where a minor second peak of radioactivity was observed at

Table 2
Distribution of ^{137}Cs (kBq m^{-2}) in different horizons at sampling sites (1987–2008).

Site/Horizons	cm	Year						
Skogsvallen								
		1987	1992	1994	2000	2003	2004	2007
Ah1	0–3	87.47	84.56	59.22	51.99	55.16	34.44	37.94
Ah2	3–5	1.78	4.62	23.72	19.96	30.32	18.77	26.79
Bw1	5–20	1.16[a]	2.23	5.36	10.00	27.30	18.44	31.83
Bw2	20–35	–	0.30[b]	0.50[b]	0.23	0.36	0.85	1.49
BC	35–50	–	–	–	0.09	0.12	0.38	0.56
∑ (mean)		90.4	91.7	88.8	82.3	113.3	72.9	98.6
Möjsjövik								
		1987	1994	1998	2000	2002	2004	2007
Hap	0–20	71.45	55.68	48.05	56.91	75.67	72.30	68.42
He	20–50	–	0.29[b]	0.54[c]	1.39	2.32	1.63	2.25
∑ (mean)		71.5	56.0	48.6	58.3	78.0	73.9	70.7
Hille								
		1987	1990	1994	2000	2002	2005	2007
Hap	0–25	169.66	138.54	202.91	245.69	175.63	188.70	181.50
He	25–33	–	–	–	2.34[d]	0.96[g]	6.57[g]	5.35[g]
Hi	33–47	–	–	–	0.54[e]	0.47[h]	5.43[h]	3.80[h]
2Cg	47–55	–	–	–	0.21[f]	0.17[i]	0.20[i]	0.66
∑ (mean)		169.7	138.5	202.9	248.8	177.2	200.9	191.3
Ramvik								
		1987	1994		2000		2003	2007
Ah1	0–7	46.22[j]	39.93[l]	–	47.98	–	12.72	48.83
Ah2(Ap)	7–23	1.81[k]	5.32[m]	–	23.45[n]	–	10.29[p]	27.08
Bw	23–34	–	–	–	0.17[d]	–	0.50[q]	0.75[q]
CBg	34–55	–	–	–	0.21[o]	–	0.02[r]	0.07[o]
∑ (mean)		48.0	45.2	–	71.8	–	23.5	76.7
Hammarstrand								
		1989	1994		2000		2003	2007
Ah1	0–6	39.27[l]	19.83[l]	–	48.52	–	28.91	34.51
Ah2(Ap)	6–20	3.21[m]	4.12[m]	–	9.10	–	6.60	5.59
Ah3	20–30	0.17[b]	0.39[b]	–	0.11	–	0.20	0.01
C	30–50	–	–	–	0.38	–	0.06	0.00
∑ (mean)		42.6	24.3		58.1		35.8	40.1
Stora Blåsjön								
		1989	1995		2000		2003	2007
Oi	0–2	14.98	7.68	–	26.50	–	16.48	25.61
Ah	2–4	7.43	8.23	–	10.55	–	10.60	11.61
Ah/E/B(Ap)	4–15	3.95	3.06	–	5.49	–	3.89	5.99
Bs(Bw)	15–35	4.65[s]	1.11[s]	–	0.54	–	0.56[s]	0.82
Bc	35–60	–	–	–	0.24	–	–	0.06
∑ (mean)		31.0	20.1		43.3		31.5	44.1
Blomhöjden								
		1990	1994		2000			2007
Oi	0–2.5	9.03[t]	5.61[t]	–	8.80	–	–	5.95
Oe	2.5–5	8.73[u]	3.53[u]	–	4.72	–	–	5.70
E	5–10	1.40	0.29	–	1.71	–	–	1.87
Bs1	10–17	1.02[v]	0.31[v]	–	1.01[w]	–	–	0.81
Bs2	17–30	1.02[s]	0.24[s]	–	0.74[x]	–	–	0.49[x]
∑ (mean)		21.2	10.0		17.0			14.8
Åsenmossen								
					2000		2003	2008
Hi1	0–15	–	–	–	28.72	–	23.26	28.10
Hi2	15–25	–	–	–	1.33	–	1.06	1.70
He1	25–35	–	–	–	0.97	–	0.91	1.00
He2	35–45	–	–	–	0.25	–	0.73	1.35
He3	45–55	–	–	–	–	–	0.83	1.27
∑ (mean)					31.3		26.8	33.4

[a] 5–15 cm; [b] 20–25 cm; [c] 20–35 cm; [d] 25–35 cm; [e] 35–45 cm; [f] 45–55 cm; [g] 25–32.5 cm; [h] 32.5–47.5 cm; [i] 47.5–55.0 cm; [j] 0–7.5 cm; [k] 7.5–25.0 cm; [l] 0–5 cm; [m] 5–20 cm; [n] 7-25.0 cm; [o] 35–55 cm; [p] 7–22.5 cm; [q] 22.5–35 cm; [r] 35–40 cm; [s] 15–25 cm; [t] 0–2 cm; [u] 2–5 cm; [v] 10–15 cm; [w] 10–17.5 cm; [x] 17.5–30 cm.

depths between 10 and 15 cm. In 1987, the migration rates of the radionuclide were lower at Skogsvallen (0.13 cm yr$-^1$), moderate at Möjsjövik (0.17 cm yr$-^1$) and Ramvik, (0.19 cm yr$-^1$), and higher at Hille (0.23 cm yr$-^1$) (Figs. 3.1–3.4).

Two years later (1989), similar patterns were observed in the mineral soil profiles at Hammarstrand and Stora Blåsjön, with the peak of radioactivity located at depths of 0–4 cm, accounting for about 77 % of total radioactivity and migration rates of 0.26 cm yr$-^1$ and 0.42 cm yr$-^1$, respectively (Figs. 3.5, 3.6). The migration depth of the radionuclide was 2.43 cm at Hammarstrand and 5.09 cm at Stora Blåsjön, where a slight second peak of radioactivity was recorded at a depth of 15–20 cm.

By 1990 (year three), about 80 % of the radioactivity was in deeper soil layers (3–5 cm), with migration rates of 0.27 cm yr$-^1$ at Hille and 0.37 cm yr$-^1$ at Blomhöjden (Figs. 3.5, 3.7), corresponding to migration depths of 2.65 cm and 3.37 cm, respectively.

After five years (1992), samples from the Skogsvallen site indicated that approximately 84 % of the total activity remained at a depth of 0–2 cm, with a migration rate of 1.42 cm yr$-^1$ and a migration depth of 1.42 cm (Fig. 3.1).

The intermediate period (1994–2003; year 8–17). From 1994 to 2003, radionuclide distribution peaks were evident in the upper 0–5 cm of mineral and organic soils at several sites, with 80–86 % of total activity found in Skogsvallen, Ramvik, Stora Blåsjön, and Möjsjövik (Fig. 3a, 3d, 3f, 3b). Migration rates varied from 0.16 cm yr^{-1} in 2000 at Stora Blåsjön to 0.47 cm yr^{-1} in 2003 at Ramvik, leading to deeper penetration of ^{137}Cs at Ramvik, which increased from 3.8 cm in 1994 to 7.6 cm in 2003. At Blomhöjden, 80 % of radionuclide activity remained in the upper 0–4 cm, while Hammarstrand saw a shift in peak radioactivity depth from 0–5 cm layer in 1994 to 1–7 cm in 2003 (Figs. 3.7, 3.5). Additionally, a

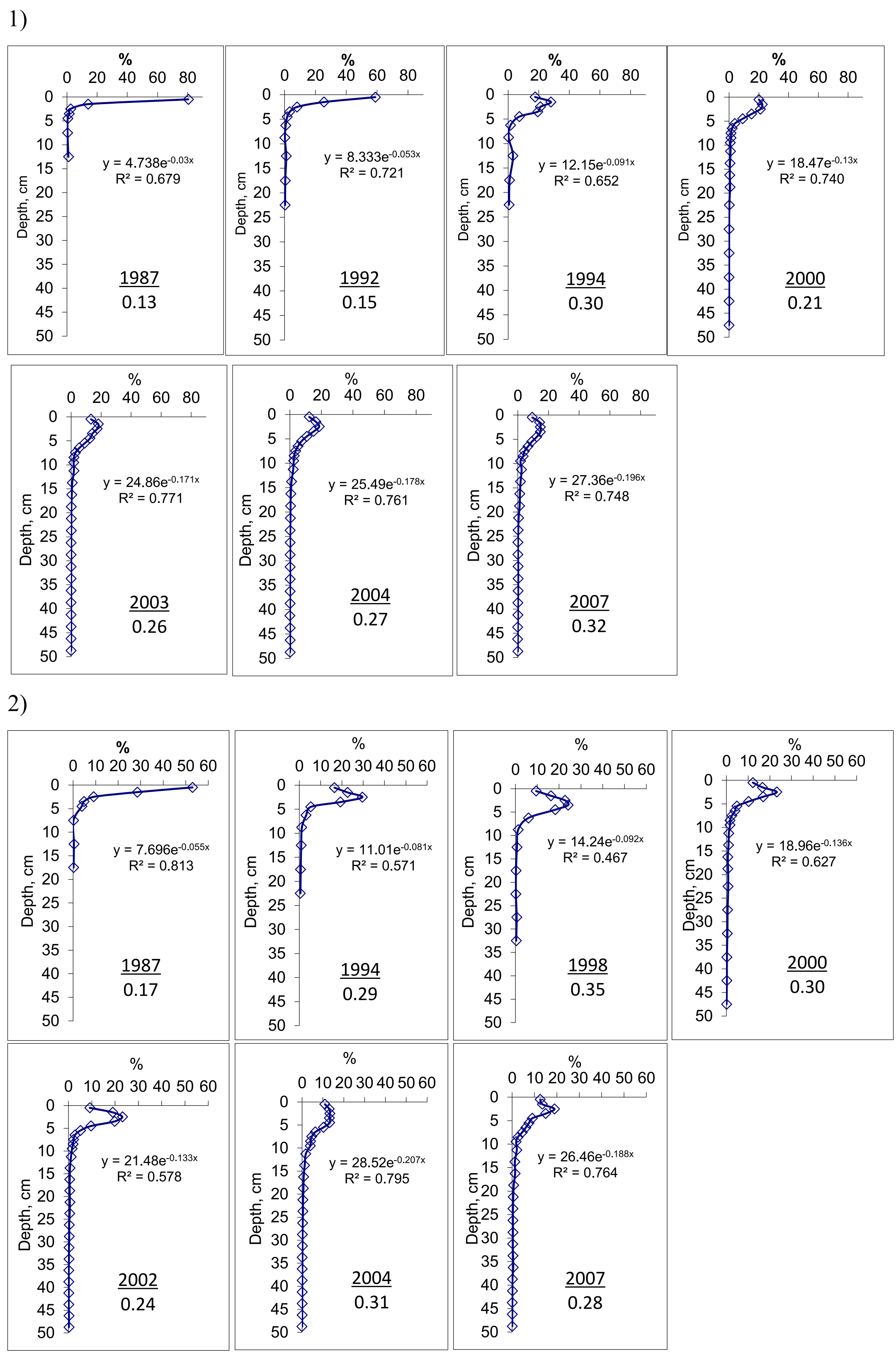


Fig. 3. Distribution of ^{137}Cs, % (Bq/m^2)/cm in soil profiles. Numerator – year, denominator – migration rate, cm year^{-1} (0–20 cm). 1) Skogsvallen; 2) Möjsjövik; 3) Hille; 4) Ramvik; 5) Hammarstrand; 6) Stora Blåsjön; 7) Blomhöjden, and 8) Åsenmossen.

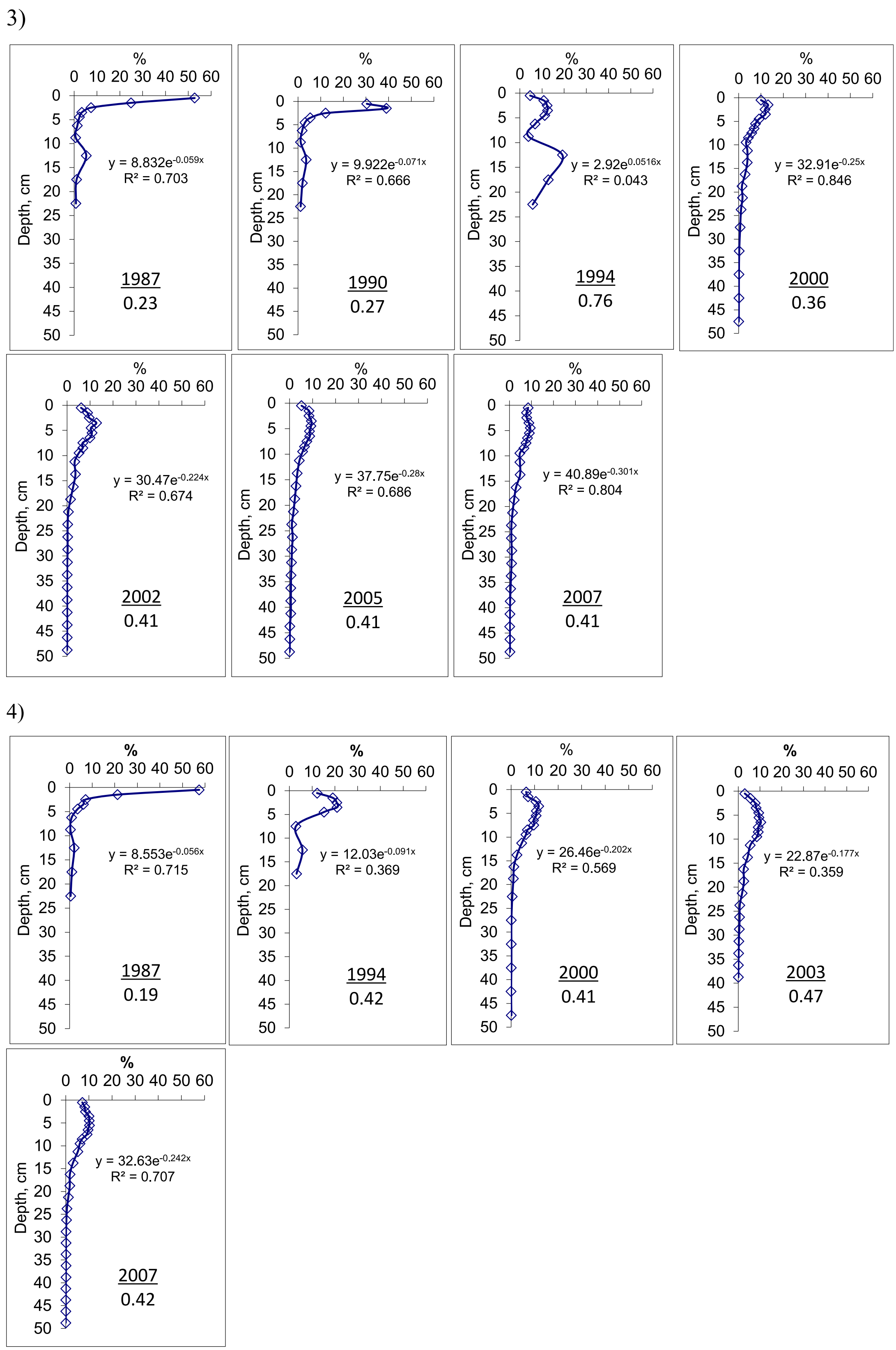
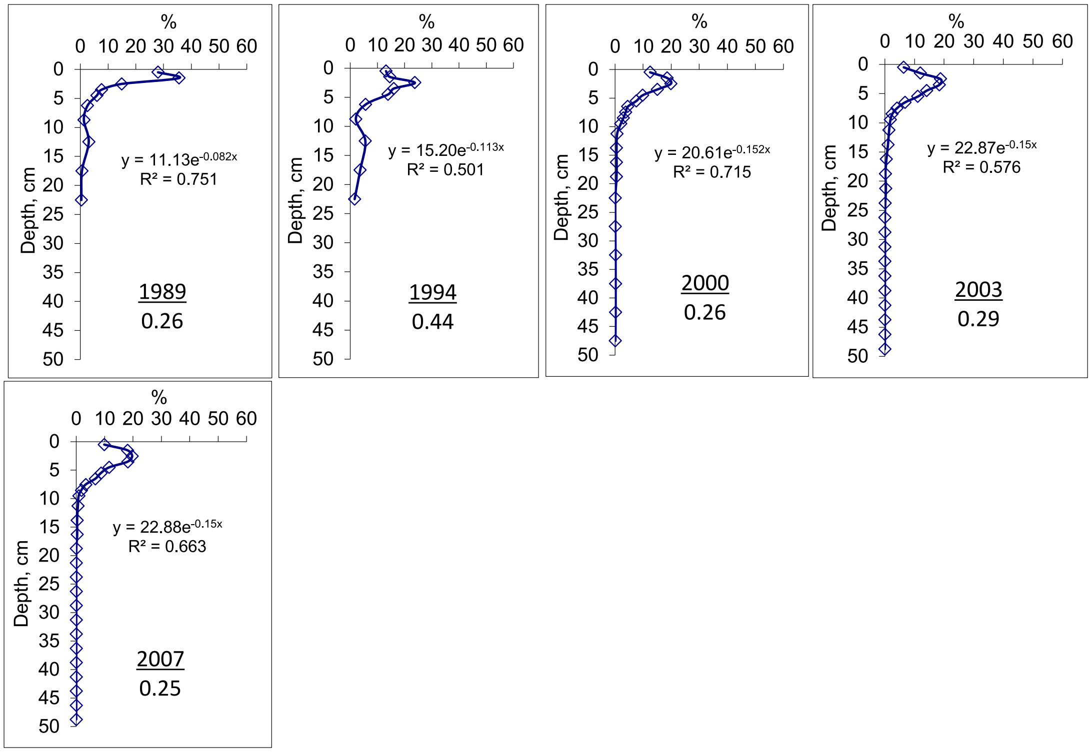


Fig. 3. (*continued*).

secondary maximum of ^{137}Cs activity at Hille, present at 10–20 cm in 1994, gradually diminished by 2000 (Fig. 3.3).

The later period from 2004 to 2008 (Year 18–22). At five of the eight sites (Skogsvallen, Möjsjövik, Hammarstrand, Stora Blåsjön, and Blomhöjden), approximately 80 % of total ^{137}Cs activity was found in the upper 0–6 cm layers, regardless of soil type (Figs. 3.1, 3.2, 3.5, 3.6, 3.7). Migration rates at these sites ranged from 0.17 cm yr^{-1} at Stora Blåsjön to 0.32 cm yr^{-1} at Skogsvallen, averaging 0.26 cm yr^{-1}, with an

5)



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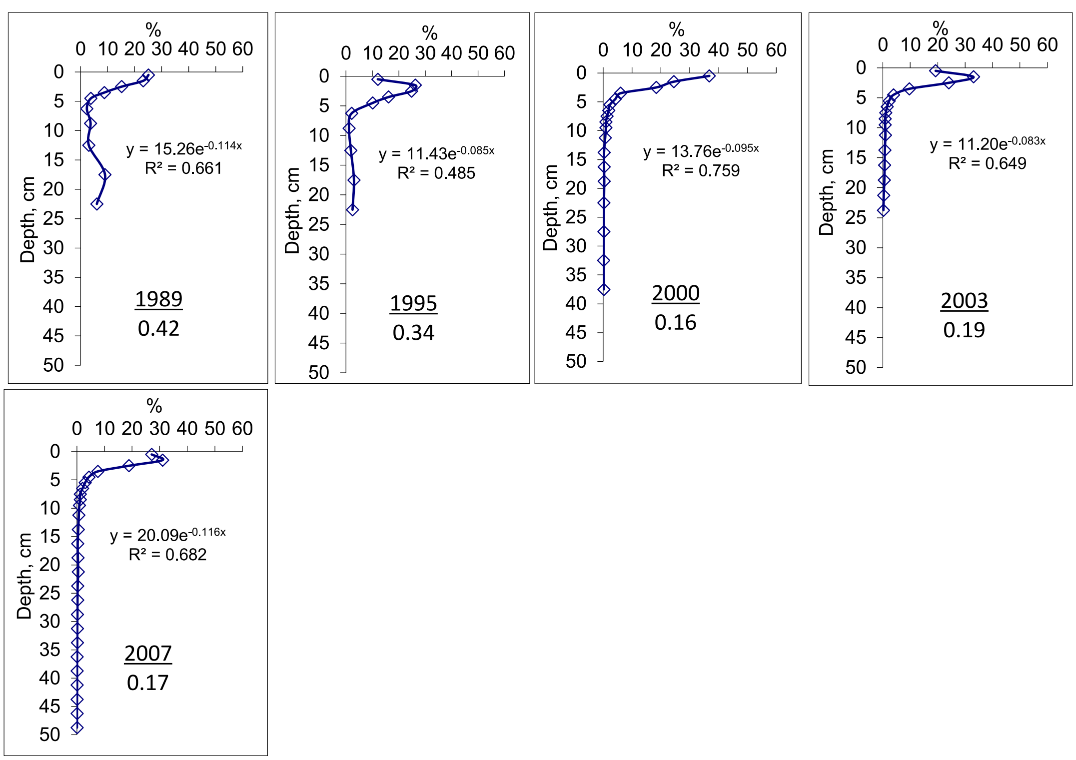

Fig. 3. (*continued*).

average migration depth of 4.3 cm (range 2.7–4.9 cm). At Hille and Ramvik, 75–78 % of activity was within the 0–9 cm layers, with average migration rates and depths of 0.41 cm yr^{-1} and 7.5 cm, respectively. At Åsenmossen in 2008, two peaks were observed, with 64 % of radioactivity at 0–7 cm and 20 % at 10–15 cm, a migration rate of 0.37 cm yr^{-1}, and an average depth of 8.9 cm (Fig. 3.8).

The migration rate of ^{137}Cs was analysed across the different soil profiles categorized by type: mineral soils (Skogsvallen, Ramvik, Hammarstrand, Stora Blåsjön, Blomhöjden), and organic soils (Möjsjövik, Hille, Åsenmossen). In both mineral and organic soils, the migration rate

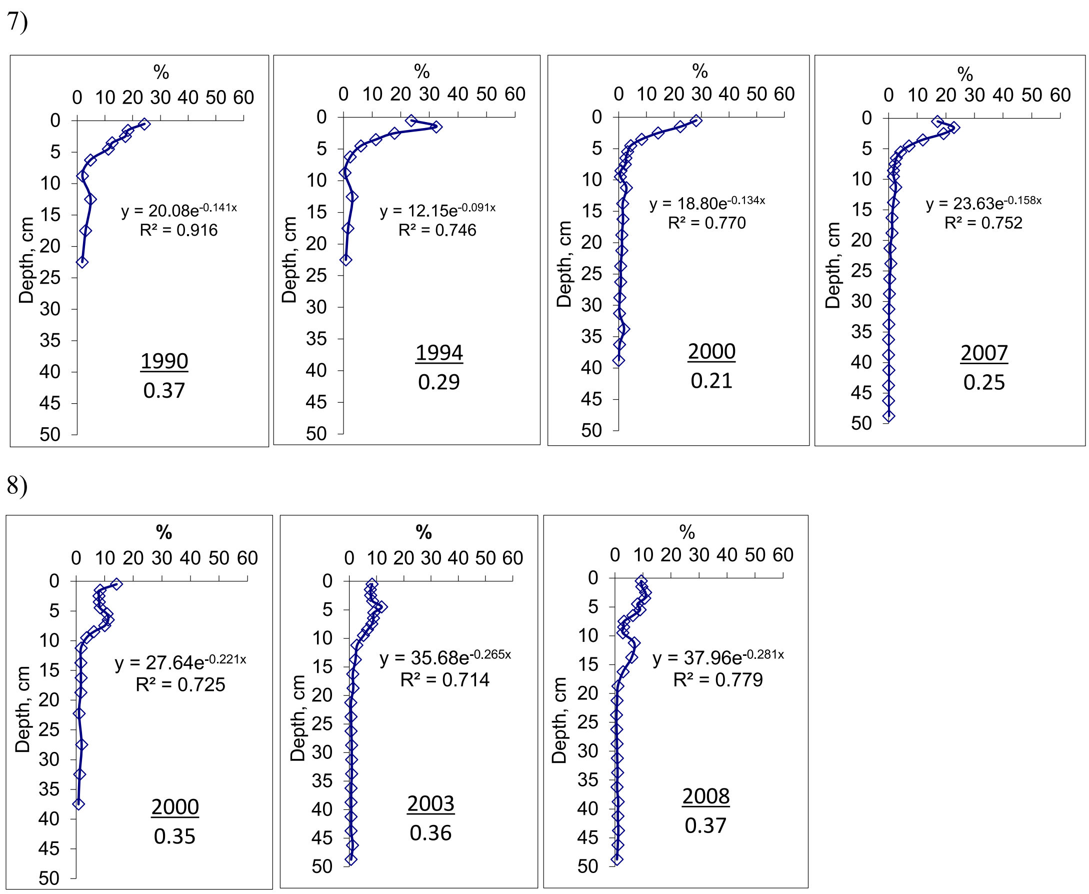

Fig. 3. (*continued*).

of ^{137}Cs remained stable over time (Fig. 4), averaging 0.28 and 0.35 cm yr^{-1} respectively.

The distribution of the radionuclide in the undisturbed top layers followed an approximately exponential pattern, with R^2 values ranging from 0.36 to 0.92 (Figs. 3.1–3.8), except the Hille site in 1994 (Fig. 3.3). According to the Mann–Whitney *U* test results (Table 3), the distribution of ^{137}Cs in soil profiles was similar in 1987 but exhibited differences in 2000 and 2007.

In 2000, the migration depth of ^{137}Cs at Skogsvallen (3.1 cm) was significantly shallower than at Möjsjövik, Hille, Hammarstrand, Blomhöjden and Åsenmossen, but deeper than at Stora Blåsjön. The peat soils at Möjsjövik and Hille, along with the loam soil at Ramvik, had deeper ^{137}Cs locations compared to the sandy loam at Stora Blåsjön (3.7 cm). By 2007, ^{137}Cs locations at Skogsvallen and Möjsjövik were significantly shallower than those at Hammarstrand and Stora Blåsjön, with Hammarstrand (3.5 cm) having shallower distribution compared to Skogsvallen, Möjsjövik, Hille, and Ramvik, while Stora Blåsjön had the shallowest location (2.7 cm) (Table 3). Meanwhile the migration depth within soil profiles can vary significantly due to differences in soil type, thickness, and bulk density, among other factors.

To summarize, the migration depth of ^{137}Cs in soil profiles, the data were divided into three periods: early (1987–1992), intermediate (1994–2003), and late (2003–2007), and compared across two groups of soil types: mineral and organic soils (Table 4).

^{137}Cs distribution in soil layers changed with time after fallout. During the early period (1–6 years post-fallout), ^{137}Cs was primarily found in the upper soil layers (Table 4). At this time the migration depth was not significantly different between the mineral and organic soils. However, in the intermediate (7–17 years post-fallout; $P = 0.064$) and later (18–21 years post-fallout; $P = 0.039$) period, ^{137}Cs was found at significantly deeper layers in organic soils (Table 4). After 20 years, ^{137}Cs was found at significantly deeper layers in mineral soils as compared to the initial phase whereas in organic soils the average migration was deeper already in the intermediate period (Table 4).

There was no significant relationship between soil clay content in mineral soils and the average migration depth of the radionuclide during the study period ($P = 0.423$).

4. Discussion

The results show that approximately 20 years after deposition, the pattern of ^{137}Cs distribution within the soil profile is generally similar across all studied soils, including mineral silty clay, silty loam, loam, sandy loam, and organic soils. The distribution of ^{137}Cs in the profiles of five undisturbed mineral soils and three organic soils shows very slow downward migration, consistent with earlier findings in Sweden (Rosén et al., 1999; Almgren & Isaksson, 2006). The radionuclide primarily remains in the upper soil layers, continuing to be a source of exposure, e.g. through plant or fungi uptake. In 1987, about 80 % of radionuclide activity was in the top 0–2 cm of soil, but by 2007, this peak shifted to the 0–6 cm layer across all sites. Sigurgeirsson et al. (2005) also observed this pattern in coarse-grained soil with 2–5 % clay content and less than 1 % organic matter, indicating that even small amounts of clay can inhibit radionuclide migration to deeper layers.

The strong retention and slow migration of ^{137}Cs in mineral soils is likely due to the presence of 2:1 phyllosilicates like illite, vermiculite (Cremers et al., 1988; Fuller et al., 2015), and kaolinite (Szabó et al., 2012). The deeper migration of ^{137}Cs in organic soils is likely related to their generally lower adsorption of ^{137}Cs (Sanchez et al., 1999; Chibowski & Zygmunt, 2002; Tatsuno et al., 2020). However, the minimal radioactivity migrated below 5–6 cm is probably linked to the non-specific adsorption of ^{137}Cs to the ion exchange complex of organic

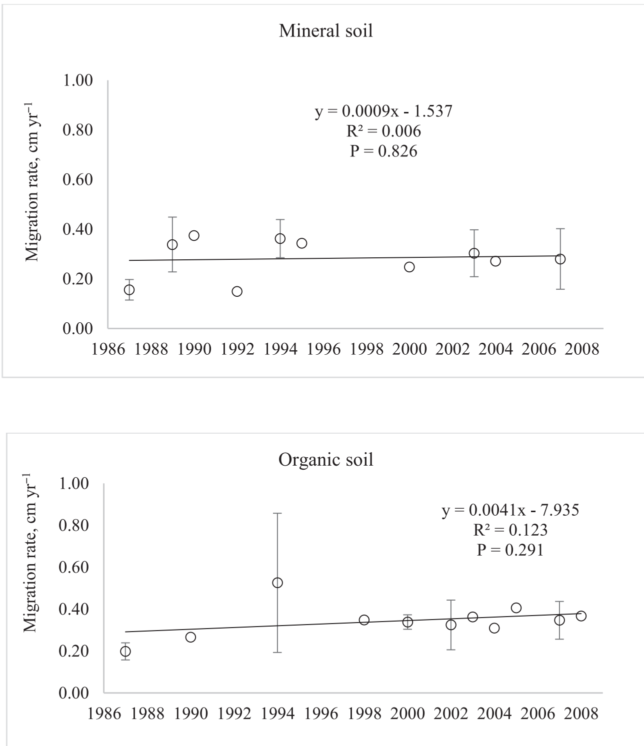

Fig. 4. Migration rate of ^{137}Cs (mean ± SD) in different soils, cm yr^{-1}: mineral soil (Skogsvallen, Ramvik, Hammarstrand, Stora Blåsjön and Blomhöjden); organic soil (Möjsjövik, Hille, Åsenmossen). The P indicates the probability of ^{137}Cs migration rate change over time.

matter (Rigol et al., 2002).

The involvement of earthworm populations in radionuclide turnover may also influence the distinct distributions and migration rates of ^{137}Cs in grassland soils over time, highlighting the significance of this transport mechanism (Jarvis et al., 2010). This study cannot estimate the extent of earthworm involvement in ^{137}Cs distribution; however, anecic earthworms in finer-textured undisturbed soils may create a linear ^{137}Cs distribution with depth (McCabe et al., 1991), thereby affecting its migration rate.

The retention and slow migration of ^{137}Cs in undisturbed grassland soils are likely influenced by plant roots. Studies indicate that roots of e.g. clover (*Trifolium* spp.) and bean (*Vicia* spp.) contain significant amounts of ^{137}Cs, holding 16–22 % of the total radionuclide in the plant (Ramadan et al., 2021). This results in higher ^{137}Cs concentrations in the root zone due to uptake and redistribution during plant decay. Plant roots can modify soil structure and hydraulic properties, affect the movement of water and solutes (Xiao et al., 2024) and potentially redistribute ^{137}Cs in the soil.

At the Ramvik (sandy loam) and Hammarstrand (silty loam) sites, more than 80 % of radioactivity in 2007 were detected in slightly deeper soil horizons (0–9 cm). The sites had comparable organic carbon content (4 %) and relatively high silt and clay content (54 % and 66 %) in the Ah1 horizon. The deeper penetration of the radionuclide at these sites is likely due to a second peak of ^{137}Cs activity observed from 1987 to 1994. Four of the five mineral soils (Ramvik, Hammarstrand, Stora Blåsjön, and Blomhöjden) and one of the three organic soils (Hille) showed a notable second maximum of activity in the 10–20 cm layers during that period, which gradually diminished in later years. The cause of the second peak observed in those soils is still unclear. This may be attributed to the Chornobyl fallout in Sweden, primarily occurring through

Table 4
^{137}Cs migration depth, mean and standard deviation, within mineral and organic soil profiles during three periods after the Chornobyl fallout: 1–6 years, 7–17 years and 18–21 years.

Site	Depth, cm 1987–1992	1994–2003	2004–2008
Mineral soil			
Skogsvallen, Ramvik, Hammarstrand, Stora Blåsjön, Blomhöjden	2.49 ± 1.54 (6)^{n}	3.97 ± 1.40 (14)	4.37 ± 1.27 ^{*a} (6)
Organic soil			
Möjsjövik, Hille, Åsenmossen	2.08 ± 0.66 (3)	5.51 ± 1.99 **(9)	6.98 ± 1.88 ^{**b} (5)

^{n} in brackets number of observations, sites/years; * $p < 0.05$; ** $p < 0.01$ indicate differences between the time periods. Different small letters (a, b) indicate differences between mineral and organic soils.

Table 3
P-values according to Mann–Whitney *U* test (figures in cursive are < 0.05) of the difference between the migration depths (X) of ^{137}Cs in soil profiles in selected years.

Sites	Depths of ^{137}Cs location, cm	Möjsjövik	Hille	Ramvik	Hammarstrand	Stora Blåsjön	Blomhöjden	Åsenmossen
		1987						
Skogsvallen	0.89	0.603	0.157	0.223	n/d*	n/d	n/d	n/d
Möjsjövik	1.36		0.505	0.689	n/d	n/d	n/d	n/d
Hille	2.22			0.678	n/d	n/d	n/d	n/d
Ramvik	1.76				n/d	n/d	n/d	n/d
		2000						
Skogsvallen	3.06	***0.036***	***0.016***	0.072	***0.006***	***0.006***	***0.043***	***0.001***
Möjsjövik	4.24		0.127	0.579	***0.000***	***0.016***	0.521	***0.006***
Hille	5.80			0.969	***0.000***	***0.012***	0.056	0.526
Ramvik	5.93				***0.000***	***0.019***	0.554	0.260
Hammarstrand	3.93					***0.002***	***0.000***	***0.000***
Stora Blåsjön	2.46						***0.013***	***0.000***
Blomhöjden	4.58							***0.006***
Åsenmossen	6.40							
		2007						
Skogsvallen	5.14	0.763	0.023	0.929	***0.019***	0.056	0.615	***0.006*****
Möjsjövik	4.90		***0.018***	0.964	***0.012***	***0.007***	0.504	***0.001*****
Hille	7.88			0.197	***0.000***	***0.000***	***0.004***	0.615**
Ramvik	6.38				***0.041***	0.185	0.459	0.077**
Hammarstrand	3.54					0.238	0.081	***0.000*****
Stora Blåsjön	2.71						0.077	***0.000*****
Blomhöjden	4.15							***0.004*****
Åsenmossen	8.85							

*n/d stands for no data.
** for the year 2008.

wet deposition linked to precipitation (Persson et al., 1986), when the readily soluble 1986 activity rapidly infiltrated the soil, exceeding the long-term rate of caesium migration (Schimmack et al., 1989). This could lead to a second peak in ^{137}Cs activity in later years. Research also shows a strong correlation between the amount of ^{137}Cs in deeper soil layers and total annual precipitation (Graham & Simon, 1996; Sigurgeirsson et al., 2005). Alternatively, secondary peaks may appear locally in some replicate samples where the standard deviation of the ^{137}Cs inventory within individual soil layers is high. The second peak may have influenced radionuclide migration speed and depth, potentially due to the soil structure and the wet deposition process, which could have allowed some of the water-soluble radioactivity to penetrate deeper layers immediately after deposition, although this hypothesis cannot be conclusively proven within this study.

A similar second peak in radioactivity was observed at a comparable depth in peat soil at Åsenmossen in 2008, likely due to the dominance of sphagnum moss, which has the highest ^{137}Cs activity in the capitula or subapical segments, suggesting ongoing relocation of elements to the actively growing apical part (Vinichuk et al., 2010).

The average radioactivity migration rate across various sites was 0.31 cm y$-^{1}$ (range: 0.127–0.76 cm y$-^{1}$), peaking at 0.35 cm y$-^{1}$ in organic soils (Möjsjövik, Hille, Åsenmossen). In mineral soils, the radionuclide migration rates were 0.28 cm y$-^{1}$ as an average with a range between 0.13–0.47 cm y$-^{1}$. These findings generally align with previous studies, being somewhat higher than Arapis et al. (1997), Almgren & Isaksson (2006) and Ajayi et al., (2007) but lower than Ivanov et al. (1997) and Forsberg & Strandmark (2001). The ^{137}Cs migration rates varied by soil type, with no reduction over time in both mineral and organic soils.

In this work, we used "average depth" as the calculation method because the original ^{137}Cs data (Bq m$-^{2}$ / cm) exhibited fewer outliers in later years following fallout. Early-year radionuclide distributions in soil profiles (Rosén et al., 1999) were uneven, making median depth more appropriate as it is less sensitive to extreme values. This study builds upon the work of Rosén et al. (1999), employing similar methods. Results are comparable to other studies using similar methods (e.g., Arapis et al., 1997; Forsberg & Strandmark, 2000) or those utilizing alternative approaches (e.g., Almgren & Isaksson, 2006), when the estimation of the caesium transport in the soil was based on the convection and diffusion equation.

5. Conclusions

The main finding of this research is that radiocesium remains concentrated in the upper 0–10 cm layers of undisturbed grassland soil more than 20 years after the fallout indicating that the radionuclide is still accessible for absorption by plant roots.

Long-term observations also allowed the calculation of key parameters, such as the migration rate and depth of radiocesium distribution. In an intermediate (7–17 years) and long (18–21 years) time perspective, the average ^{137} Cs migration depth was deeper in organic soils (7.0 cm) as compared to mineral soils (4.4 cm), having more specific binding sites for ^{137}Cs.

Some soils initially showed a secondary peak of radioactivity at 10–20 cm, which diminished over time. This peak may have affected radionuclide migration speed and depth, potentially due to soil structure and wet deposition, which could allow rapid penetration of water-soluble radioactivity.

CRediT authorship contribution statement

Klas Rosén: Writing – review & editing, Data curation, Conceptualization. **Ingrid Öborn:** Writing – review & editing, Validation, Conceptualization. **Mykhailo Vinichuk:** Writing – review & editing, Writing – original draft, Visualization, Validation, Formal analysis.

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.geoderma.2025.117479.

Data availability

Data will be made available on request.

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