Journal of Environmental Radioactivity 282 (2025) 107621

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Long-term transfer of ^{137}Cs in sensitive agricultural environments after the Chornobyl fallout in Sweden

Mykhailo Vinichuk [a,b,*], Magnus Simonsson [b], Maja Larsson [b], Klas Rosén [b]

[a] Department of Ecology and Environmental Protection Technologies, Zhytomyr Polytechnic State University, P.O. Box 10005, Zhytomyr, Ukraine
[b] Department of Soil and Environment, Swedish University of Agricultural Sciences, Box 7070, 750 07, Uppsala, Sweden

ARTICLE INFO

Keywords:
Grass
Pasture
^{137}Cs
Soil
Clay
Sand

ABSTRACT

In this study, the long-term transfer of ^{137}Cs from soil to grass on Swedish farms and fields, heavily contaminated after the 1986 radioactive fallout, was investigated. The study spans over 8–14 years, beginning in June 1986, and covers various soil types and agricultural practices. The transfer of ^{137}Cs from soil to grass was highly variable, with transfer factors ranging from 1.0×10^{-5} to 0.357 m^2 kg^{-1}. Higher values were observed on fields with sandy loam, loamy sand, and organic soils, and lower values on fields with a high clay content. The transfer of ^{137}Cs to grass generally exhibited an exponential decline across the majority of fields over the years. The rate of decrease was most pronounced in clay loam and silty loam soils, while it was least evident in sandy loam, sandy soils, and peat soils. The soil properties and farming practices were more important for ^{137}Cs uptake than the initial deposition density. The transfer factor had a negative correlation with soil pH, clay, and fine silt content. No significant relationships were found with other soil variables, such as soil organic matter content and plant available potassium concentration. The median effective half-life of ^{137}Cs in the grass was 4.5 years, with a range of 2–18 years. The uptake of ^{137}Cs by plants did not correlate with the potassium concentration in grass tissues; however, the activity concentration of ^{137}Cs in grass correlated negatively with the potassium content in the plants grown on fields with high deposition levels.

1. Introduction

After an accidental release of radiocaesium (^{137}Cs) into the environment, the isotope enters the ecosystem through similar uptake mechanisms as potassium. As it moves through the food chain from soil to human consumption, ^{137}Cs can accumulate in the human body, posing serious health risks. To estimate the radiation dose received by humans and mitigate contamination in the event of a future accidental release, mathematical models are used. These models rely on transfer factors, which describe the movement of specific radionuclides from soil to crops, to predict radionuclide concentrations in foodstuffs (Yasuda, 1995). The acquisition of additional knowledge in the field is crucial for designing and implementing effective agricultural countermeasures following a radioactive fallout event similar to the Chornobyl accident.

Extensive research has been conducted on the transfer of ^{137}Cs from soil to agricultural crops following the Chornobyl and Fukushima nuclear accidents, revealing significant variability in transfer factors (Rosén et al., 1998; Tsukada and Hasegawa, 2002; Uchida and Tagami, 2007; Sarap et al., 2015; Yoshikawa et al., 2020). Immediately after radionuclide deposition, crop contamination is primarily influenced by factors such as fallout type, plant development stage, rainfall, and the chemical form of the radionuclide (Pröhl, 2009). Over time, uptake by roots in the soil becomes the primary pathway of crop contamination. The uptake is dependent on radionuclide properties, crop types, soil characteristics, and climatic conditions (Coughtrey and Thorne, 1983). Long-term studies on ^{137}Cs transfer from soil to crops are limited, although some studies dealing with the long-term trend in plant availability of radionuclides have been published (Krouglov et al., 1997; Brimo et al., 2020).

The variability of ^{137}Cs transfer rates limits the effectiveness of a single concentration factor for predicting radionuclide uptake by crops from soil (Ng, 1982). However, the uncertainties can be reduced by considering the dominant crop and soil types in the area, as well as the effects of various soil properties on radionuclide transfer. Following the 1986 Chornobyl nuclear power plant accident, ^{137}Cs contamination was highly variable in Sweden and the deposition was correlated with

* Corresponding author. Department of Ecology and Environmental Protection Technologies, Zhytomyr Polytechnic State University, P.O. Box 10005, Zhytomyr, Ukraine.

E-mail address: Mykhailo.Vinichuk@slu.se (M. Vinichuk).

https://doi.org/10.1016/j.jenvrad.2025.107621
Received 6 November 2024; Received in revised form 18 January 2025; Accepted 19 January 2025
Available online 23 January 2025

rainfall patterns after the accident (Isaksson et al., 2000). Aerial measurements conducted between May and October 1986 revealed ^{137}Cs deposition range up to 70 kBq m^{-2} in Uppsala county, 20 to >80 kBq m^{-2} in Gävleborg, 10 to >60 kBq m^{-2} in Västernorrland, and 5 to >80 kBq m^{-2} in Jämtland (Fig. 1).

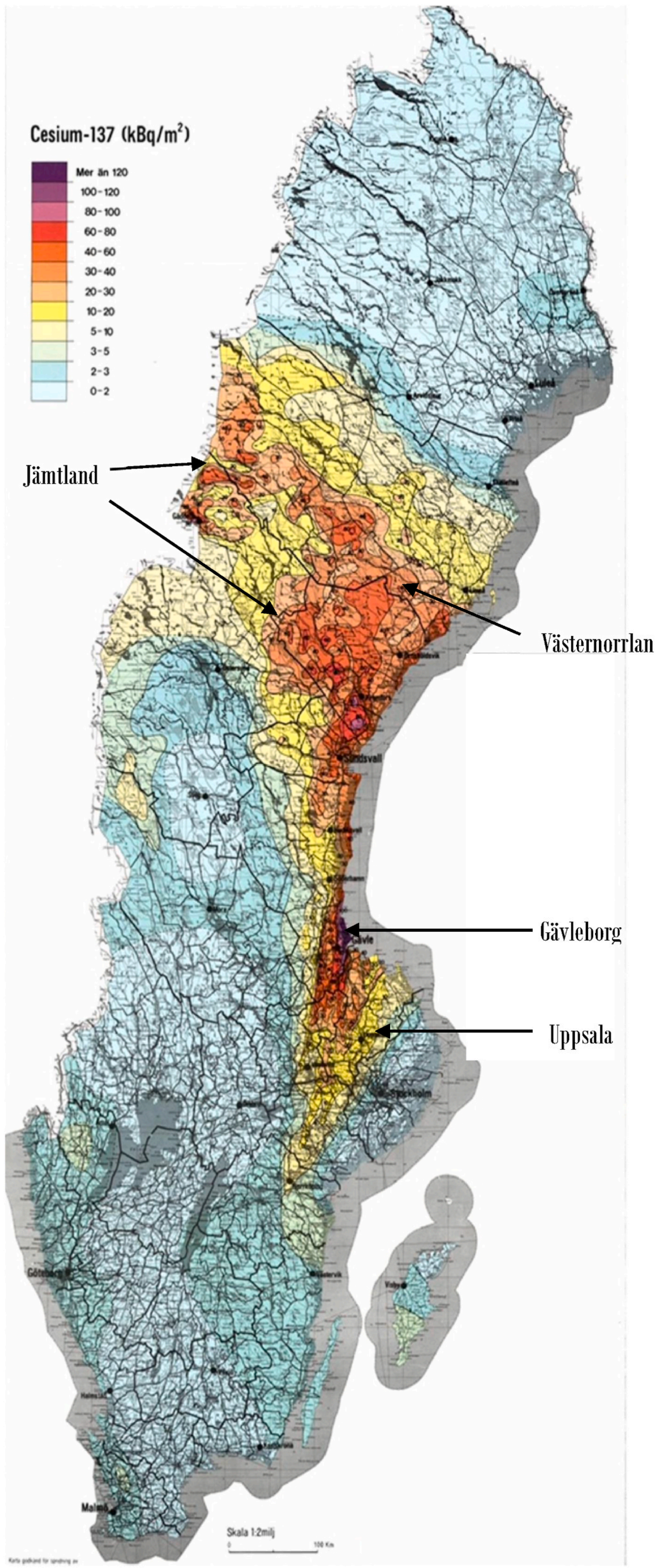

Fig. 1. Deposition density of ^{137}Cs from Chornobyl, based on inflight measurements taken from May to October 1986 in Sweden (SGAB, 1986).

Whereas previous publications of Rosén et al. (1996a,b, 1998) were restricted to the early years after the fallout (1986–1996), the present paper gives an overview of data from these most affected counties on a longer term, 1986–2007. The data on ^{137}Cs activity concentrations in soil and grass were collected and analysed. The objectives of the present study were to investigate (1) the long-term pattern of ^{137}Cs transfer from soil to grass on various soil types, when the dynamics of ^{137}Cs within the soil-grass system have attained an apparent steady state, (2) the influence of soil properties, including pH, organic matter content, and texture, on ^{137}Cs transfer from soil to grass, (3) the effect of readily available soil potassium on ^{137}Cs transfer from soil to grass, and (4) whether the ^{137}Cs activity concentration in plants is correlated with the concentration of potassium and calcium in the grass tissues.

2. Materials and methods

2.1. Fields and sampling area

The fieldwork was conducted over a period of 21 years, from 1986 to 2007 and samples were taken on 29 fields in the counties of Uppsala, Gävleborg, Västernorrland, and Jämtland. Throughout the study period, the fields remained under respective farmer's practice. Farming practices primarily relied on animal husbandry, with a main focus on ley and, occasionally, barley for fodder production. The production intensity varied, from intensive cultivation methods, involving crop rotation and mineral fertilization, to extensive cultivation focused on pasture and utilizing only animal manure as fertilizer.

As shown in Table 1, the fields were primarily used as pastures or ley, and only a few fields (X12A, X2A, X5A, X7A and Y12A) were occasionally ploughed once or twice during the years of study to a depth of about 20cm. In fields under 'intensive' cultivation (Table 1), the soil was regularly fertilized and ploughed every 4–5 years, resulting in thorough mixing of fallout into the topsoil. The first cut of grass coincided with the first mowing, primarily used for hay or silage production, whereas the second cut corresponded to regrowth used for pasture. In contrast, 'extensive' cultivation relied on permanent natural pastures, hence the soil was not ploughed. 'Moderate' cultivation fell between these two extremes (Table 1). The soil parent materials are glaciofluvial, post-glacial, or fluvial deposits with texture ranging from silty clay loam to sand; also fen peat was common in the lowland areas of Uppsala and Gävleborg.

In the latter two counties, the fields were located at 5–60 m above present sea level and 100–200 m below the postglacial marine limit. The two Uppsala fields received medium (93 and 98 kBq m^{-2}) fallout of ^{137}Cs and were under pasture. In Gävleborg, the 13 sampling sites were selected from fields with high radiocaesium content in pasture grass and hay during the initial year 1986–1987.

The fields in Västernorrland were located 150–200 m below the postglacial marine limit, within 30 km from the Bothnian coast in river valleys extending inland in a north-westerly direction. Floodplain soils on the lower terraces are characterized by a mix of sandy and clayey textures derived from alluvial deposits, while soils on upper terraces are formed in silty to clayey sediments deposited during earlier post-glacial stages when the present landscape was still submerged.

In Jämtland, three fields (Z1A, Z3bA, Z4A) are located on alluvial deposits in valleys below the postglacial marine limit, with textures ranging from loamy sand to sandy loam. The other three fields in Jämtland are located approximately 500 m above sea level in a mountainous area, on small sandy glaciofluvial deposits (Z5A, Z25A) and clayey glacial till derived from phyllite rock (Z3A).

The number of sampling occasions during 1986–2007 ranged from 8 to 14 with annual sampling of grass in the early years (1986–1996) on 2, 13, 8, and 6 fields in Uppsala, Gävleborg, Västernorrland, and Jämtland, respectively. Sampling frequency decreased over time, with collections every 2–5 years until 2007. Data on grass contamination by ^{137}Cs sampled in 1986 during the first cut in mid-June or the second cut from

Table 1
Field's properties, soil characteristics, and the transfer factors of ^{137}Cs from soil to grass ($T_{ag} \times 10^{-3}$ m^2 kg^{-1}).

Fields	Land use/ cultivation intensity	Years of sampling	^{137}Cs deposition, kBq m^{-2}[b]	T_{ag} (1st + 2nd cut)[a]		pH (H_2O)	OM, %	Clay	Silt		Sand	Soil Texture Class
				median	range			%				
								<0.002[c]	0.002–0.006	0.006–0.06	0.06–2.0	
Uppsala county												
CU11C	Pasture/ Extensive	13	93	0.49	0.01–34.6	5.7	6.2	36.5	26	19.8	17.7	Silty clay loam
CU711A	Pasture/ Extensive	10	98	6.64	2.22–67.9	5.4	–	–	–	–	–	Peat
Gävleborg county												
X1D	Ley/Extensive	10	203	1.77	0.46–357	6.1	22.1	39.2	15.2	19.5	26.2	Clay loam
X11A	Ley/Moderate	13	190	0.61	0.03–9.75	6.1	3.5	2.3	0.6	4.1	93.0	Sand
X12A	Ley/Moderate	12	95	5.79	2.36–22.3	5.8	–	–	–	–	–	Peat
X14A	Pasture/ Extensive	14	157	0.47	0.07–2.32	6.3	8.3	26.9	15.3	35.8	21.9	Silt loam
X15A	Ley/Extensive	13	179	15.1	7.96–152	5.5	8.2	5.0	1.0	8.9	85.1	Loamy sand
X15D	Pasture/ Extensive	13	149	9.27	1.38–148	5.2	11.9	5.3	1.0	10.1	83.7	Loamy sand
X18B	Pasture/ Extensive	8	83	10.9	1.43–231	4.5	–	–	–	–	–	Peat
X2A	Ley/Intensive	11	155	5.52	0.08–26.2	5.9	23.1	22.3	8.8	15.4	53.4	Sandy clay loam
X4A	Ley/Intensive	12	53	5.17	1.23–64.3	5.4	19.6	18.6	7.2	16.4	57.8	Sandy loam
X5A	Ley/Intensive	12	72	0.88	0.03–29.8	5.6	–	–	–	–	–	Peat
X7A	Ley/Moderate	9	97	1.85	0.58–72.5	5.2	–	–	–	–	–	Peat
X8B	Ley/Extensive	13	131	9.32	0.95–68.4	6.3	67.2	–	–	–	–	Peat
X9C	Ley/Extensive	10	165	5.62	0.23–104	5.4	–	–	–	–	–	Peat
Västernorrland county												
Y10A	Ley/Extensive	13	60	0.97	0.77–24.2	5.8	6.3	7.0	2.8	43.0	47.2	Loam/ sandy loam
Y12A	Ley/No data	14	28	2.02	0.45–122	–	–	–	–	–	–	Silt loam
Y15A	Ley/No data	12	46	0.78	0.27–14.8	5.5	7.0	26.6	13.6	37.6	22.2	Silt loam
Y20A	Ley/No data	14	62	0.36	0.06–3.15	6.0	6.5	24.0	17.9	19.1	39.1	Loam
Y45A	Ley/No data	14	86	0.25	0.04–3.79	5.8	6.2	26.1	16.1	32.5	25.2	Loam
Y5A	Ley/Moderate	14	22	0.55	0.01–5.98	5.5	6.0	30.4	19.3	33.0	17.3	Silty clay loam
Y55A	Ley/Moderate	12	88	0.26	0.05–2.13	5.6	3.9	6.6	3.2	30.2	60.0	Sandy loam
Y56A	Ley/Pasture/ Intensive	13	83	0.93	0.06–63.0	6.0	5.4	10.4	5.1	43.8	40.8	Loam
Jämtland county												
Z1A	Pasture/ Extensive	11	38	11.6	0.64–157	5.7	3.7	7.9	7.8	66.5	17.7	Silt loam
Z25A	Pasture/ Extensive	11	37	6.18	1.66–61.1	5.3	6.4	5.4	5.4	41.1	40.8	Sandy loam
Z3A	Pasture/ Extensive	13	44	42.0	2.64–167	5.4	4.8	4.9	3.2	40.3	51.6	Sandy loam
Z3bA	Pasture/ Extensive	12	24	6.14	0.38–70.9	5.5	3.4	3.9	1.2	27.5	67.5	Sandy loam
Z4A	Ley/Intensive	8	34	0.74	0.11–48.9	6.3	3.1	7.3	6.2	66.3	20.2	Silt loam
Z5A	Ley/Moderate	9	24	6.78	4.57–66.4	5.6	5.5	4.4	3.1	36.3	56.2	Sandy loam

[a] Over the study period for each field/farm (1986–2007).
[b] The ^{137}Cs activity concentrations were corrected to April 26, 1986.
[c] Particle size in mm.

July to October are included in Table 1 (T_{ag}, 1st + 2nd cut). However, these data were not used in future analyses, because direct fallout rather than uptake from the soil was a likely pathway of contamination of the plan.

2.2. Sampling, preparation and analyses of crops and soils

For each sampling site, a 100 m^2 land area was designated at least 100 m away from built structures, large trees, and forest edges. Within each area, soil and crop materials were randomly sampled from 1 to 5 microplots of 0.25 m^2 each. Grass was collected following local harvest practices on 1–2 sampling occasions per season, referred to as 'first cut' and 'second cut'. The grass was cut 5 cm above the soil surface, dried at 70–90 °C for 1–2 days, weighed, and then ground to <2 mm and stored in calibrated 1-L Marinelli beakers or 330-mL plastic containers. Aliquots of the grass samples were used for radiometric and chemical analyses. The content of Ca and K in plants (grass) was determined by ICP-MS after digestion in HNO_3 once in 1986.

In 1986, soil samples from each microplot were obtained by extracting 3–4 cores with a steel bore 57 mm in diameter, to a depth exceeding the expected maximum ^{137}Cs migration depth at the time of interest (usually 0–5 cm in 1986 and 0–20 cm in 2007). The core samples were pooled into a bulk sample, air-dried at 30 °C for one week, ground, and passed through a 2 mm sieve. The processed samples were then used for chemical analysis, texture determination, and estimation of ^{137}Cs deposition in 1986. Soil pH, exchangeable potassium and calcium, as well as organic matter (OM), were analysed in 0–20 cm depth in 2007. For soil sampling in 2007, a steel bore with a diameter of 21 mm was utilized. Only data from 2007 are presented in Table 1.

The texture was analysed using the hydrometer method (Day, 1965). The OM content was determined as the loss of ignition. Exchangeable calcium and potassium were extracted using ammonium acetate-lactate solution (Ca-AL and K-AL, respectively) (Egnér et al., 1960).

The activity concentrations of ^{137}Cs in soil and grass samples were measured using a computer-aided germanium detector system housed in a low-background laboratory. The relative measurement uncertainty was 1–5%. All activity concentrations reported for soil and crops are based on dry weight (d.w.). The deposition of ^{137}Cs (kBq m^{-2}) was calculated using the ^{137}Cs activity concentration in the soil cores (Bq kg^{-1}), the bulk density (g cm^{-3}) and soil depth (0–20 cm). The data on ^{137}Cs activity concentration in soil (Bq kg^{-1}) are presented in the "Supplementary material (Table S1)".

2.3. ^{137}Cs transfer to grass and its reduction with time

The transfer of ^{137}Cs from soil to grass was evaluated using the aggregated transfer factor (referred to in the text as 'transfer factor'), denoted as ^{137}Cs T_{ag} (m^2 kg^{-1}):

$$T_{ag} = \frac{A_m}{A_s} \tag{1}$$

where A_m is the activity concentration of ^{137}Cs per unit dry weight of plant material on the sampling date (Bq kg^{-1}), and A_s is the activity of ^{137}Cs (Bq m^{-2}) in soil on the same date. A_s was calculated from the deposition density of ^{137}Cs measured in 1986 ($A_{s,0}$), by using the general equation for physical decay of radionuclides, and was independent of the physical decay:

$$A_s = A_{s,0} e^{-\lambda t} \tag{2}$$

where λ, the decay constant of ^{137}Cs, y^{-1}.

Effective ecological half-life (T_{eff}), i.e. the time (years) required to achieve a 50% reduction in mean ^{137}Cs activity concentration in grass tissue, was calculated by the method of Smith et al., (1999) accordingly:

$$T_{eff} = \frac{ln(2)}{\lambda_{eff}} \tag{3}$$

where λ_{eff} is the observed rate of ^{137}Cs activity concentration decline, y^{-1}.

The rate of decrease in ^{137}Cs transfer factors in grass over the study period was calculated as a start/end ratio, specifically, the geometric means of transfer factors in the years following fallout (2nd cut in 1987 and 1st + 2nd cut in 1988) divided by the geometric means of transfer factors in the final two years of investigation (2005–2007).

2.4. Correlation analyses

Correlation and linear regression were employed to examine the relationships between (1) ^{137}Cs transfer factors and various selected soil properties, including pH, organic matter, soil texture, exchangeable potassium, and between (2) the activity concentration of ^{137}Cs in grass tissues and the concentrations of potassium and calcium in the grass.

Possible correlation of the ^{137}Cs transfer factor with the cultivation intensity was evaluated after assigning numerical scores to the cultivation intensities: 1 to 'extensive', 2 to 'moderate', and 3 to 'intensive' cultivation. The data on ^{137}Cs deposition and T_{ag} used for correlation analyses are presented in "Supplementary material (Table S2)".

3. Results

3.1. Soil characteristics and deposition density

This study is based on 29 selected fields, and spans over the period 1986–2007. The soil types and characteristics exhibited considerable variability across fields. The majority of sites were situated on peaty soils, sandy soils, and clay loam soils (Table 1).

In 1986, Gävleborg county received the highest radionuclide fallout, with an average of 135 kBq m^{-2} and a maximum of 203 kBq m^{-2}. Jämtland county had a relatively low deposition density with an average 33 kBq m^{-2} and a maximum of 44 kBq m^{-2}. The transfer factors varied significantly across the study area, with maximum values reaching 0.357 m^2 kg^{-1} (Table 1). Soil pH ranged from 4.5 to 6.3, while organic matter content varied widely between sites (4–67%) depending on soil type (Table 1).

3.2. ^{137}Cs uptake by grass

The highest transfer factors from soil to grass were observed in the first to second year following the fallout, as depicted in Fig. 2, where log-transformation has been used to reduce skewness. Over time, the transfer factors exhibit a general decline, indicating a reduction in radionuclide availability in soil for grass uptake. The fields in Fig. 2 are categorized by prevailing soil type, from clayey to sandy, and peaty. For statistical analysis, data were grouped by deposition density in 1986, and classified as low (<70), medium (70–140), or high (>140) kBq m^{-2}). A further division was done by the cultivation intensity, defined as 'intensive', 'moderate', or 'extensive'. Since the range and mean values of transfer factors were heavily affected by outliers, median rather than average values were calculated to represent each field over the whole study period. These are presented in Table 1 together with the entire intervals, including extremes.

3.2.1. ^{137}Cs uptake by grass on mineral soil

The median values of transfer factors for fields with mineral soils ranged from 4.9×10^{-4} m^2 kg^{-1} to 4.2×10^{-2} m^2 kg^{-1}. The fields with the lowest median values of transfer factors were usually found on silty clay loam and loam soils (Fig. 2a and b). On these soils, the median transfer factors on fields with low and medium deposition density were in the range 3.6–5.5 $\times 10^{-2}$ m^2 kg^{-1} (Fig. 2a and b). Even the field with the highest deposition density (X1D, 203 kBq m^{-2}), which had a soil texture of silty clay loam, had a relatively low (1.8×10^{-3} m^2 kg^{-1}) transfer factor (Fig. 2a). Notably, the transfer factors on clay loam and silty clay loam soils decreased dramatically in the years following deposition.

Radionuclide deposition in Västernorrland county was low to medium averaging 59 kBq m^{-2}, while grass uptake of ^{137}Cs was generally below 1×10^{-3} m^2 kg^{-1} (Y-fields, Fig. 2a, b and 2c). Jämtland county experienced significantly lower deposition density, with an average of 33 kBq m^{-2}, and the highest median value of transfer factors at 1.6×10^{-2} m^2 kg^{-1}, averaged for Z-fields (Table 1 and Fig. 2c). Notably, the clay content in these nutrient poor soils was relatively low, typically below 8%.

Field Z3A, characterized by sandy loam soil (Fig. 2c) and a relatively low density of ^{137}Cs deposition in 1986 (44 kBq m^{-2}), exhibited the highest median transfer factors value over the years of study, at 4.2×10^{-2} m^2 kg^{-1}. However, relatively high transfer factors values (6.8 and 5.1×10^{-3} m^2 kg^{-1}) on the same soil type were also observed on fields Z5A and X4A (Fig. 2c), which had a low (24 and 53 kBq m^{-2}) level of ^{137}Cs deposition density (Fig. 2c). The median values of transfer factors for grass grown on fields with loamy sand soils with radionuclide fallout in the range 149–190 kBq m^{-2} varied between 6.1×10^{-4} and 1.5×10^{-2} m^2 kg^{-1} (Fig. 2 d).

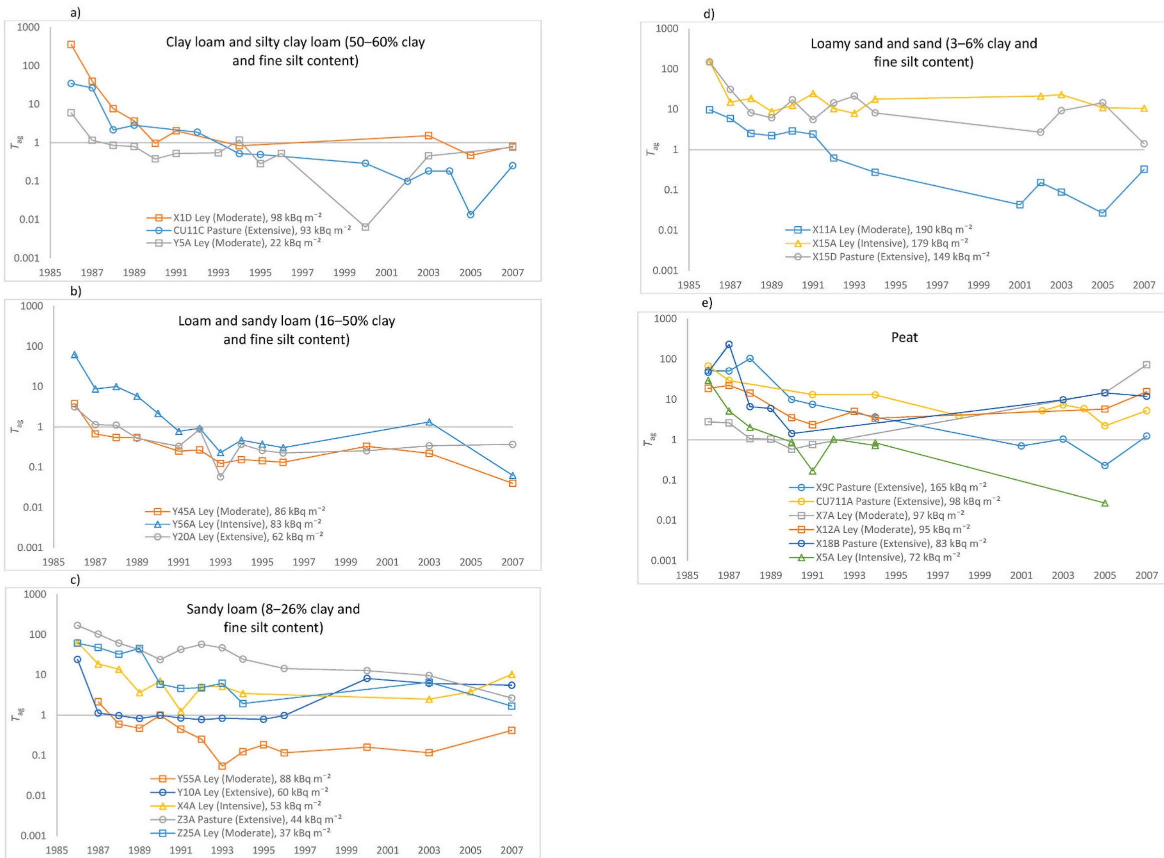

Fig. 2. The aggregated transfer factors of ^{137}Cs from soil to grass ($T_{ag} \times 10^{-3}$ m^2 kg^{-1}) on selected fields, based on an averages of 1st + 2nd cuts. Legend: CU – Uppsala; X – Gävleborg; Y – Västernorrland; Z – Jämtland, prevailing crop/land use, and deposition level of ^{137}Cs, kBq m^2 in 1986. Farming practice: circle – extensive; square – moderate; triangle – intensive.

3.2.2. *^{137}Cs uptake by grass on peat soil*

The median values of ^{137}Cs uptake by grass grown on fields with peat soil appeared to be higher compared to those on mineral soils and ranged from 8.8×10^{-4} m^2 kg^{-1} to 1.1×10^{-2} m^2 kg^{-1} (Fig. 2e). Two fields in Uppsala county that received similar low or medium fallout in 1986 exhibited significantly different transfer factor values in grass, with a median of 6.6×10^{-3} m^2 kg^{-1} on peat soil compared to 4.9×10^{-4} m^2 kg^{-1} on silty loam soil (Table 1). In fields with high deposition density (X9C, Fig. 2e), the transfer factors values were relatively high, at 5.6×10^{-3} m^2 kg^{-1}. While transfer factors on peat soils decreased significantly after 4–5 years following the fallout. Surprisingly, they increased by the end of the study period (X7A, X18B, Fig. 2e).

3.3. *^{137}Cs uptake and selected soil variables*

A correlation analysis was conducted between the average $T_{ag} \times 10^{-3}$ m^2 kg^{-1}, representing each field for the entire study period, except data from the first or second cut in 1986–1987, and selected soil characteristics.

The correlation analysis reveals no significant correlation between the transfer factor and deposition density. Soil pH exhibited a negative correlation with transfer factors ($r = -0.54$; $p = 0.003$), and the soil clay and fine-silt fractions (<0.002 mm and 0.002–0.006 mm, respectively) exhibited a moderate negative correlation with ^{137}Cs uptake by grass ($r = -0.45$ and -0.43; $p = 0.043$ and 0.053 respectively). Conversely, the coarse-silt and sand fractions (>0.06 mm) were generally not significantly correlated with ^{137}Cs uptake. The soil organic matter content and the concentration of plant-available potassium in the soil did not correlate with the transfer factors. No significant correlations were found between the ecological half-life of ^{137}Cs, T_{eff} and all studied parameters.

3.4. *The rate of ^{137}Cs transfer factors decline*

The rate of transfer factor decline over years of study was much slower on fields with sandy loam and loamy sand soils than observed in silty clay loam and loam soils, resulting in relatively high levels of ^{137}Cs even many years after the initial deposition (X15A and D, Fig. 2d). Generally, the rate of transfer factor decreased to a plateau below 1×10^{-3} m^2 kg^{-1} on silty clay loam and loams, which have up to 50–60% clay and fine silt content, as early as in 1988–1992, approximately 3–5 years after the fallout (X1D and Y5A, Fig. 2a; Y20A, Y45A and Y56A, Fig. 2b). In contrast, sandy loam soils with intermediate clay and fine silt contents (8–26%) required 12–15 years to reach the same level of ^{137}Cs uptake, regardless of the deposition density. On most of these fields, transfer factors remained above 1×10^{-3} m^2 kg^{-1} even in 2007, 21 years after the accident (X4A, Z3A and Z5A, Fig. 2c).

The transfer factors showed a significant exponential decline over the years in one-third of the fields (Table 2).

The decline ratio of T_{ag} in peat soils was variable, decreasing over the years of study in one field (CU711A) while increasing in two others (X18B and X7A) (Table 2). In contrast, the rate of T_{ag} decline throughout the study period in mineral soils, such as silty clay loam, silt loam, sandy loam, and sandy soils, was more pronounced (Table 2). Regardless of soil type and deposition density, fields with extensive farming practices had notably higher transfer factors, often three to four times greater than those found in fields with moderate and intensive farming practices.

Table 2
The functions and decline ratios of soil-to-grass transfer factors of ^{137}Cs ($T_{ag} \times 10^{-3}$ m^2 kg^{-1}) on selected fields with time, 1986–2007[a].

Fields	Soil Texture Class	Years of sampling	^{137}Cs deposition, kBq m^{-2}	Functions	P values[b]	Decline ratio
Clay loam and silty loam						
CU11C	Silty clay loam	13	93	$y = 1 + 168e^{-0.194x}$	0.001	83
X14A	Silt loam	14	157	$y = 2 + 115e^{-0.113x}$	0.001	21
Loam and loamy sandy						
Y45A	Loam	14	86	$y = 2 + 74e^{-0.086x}$	0.045	5.3
Y10A	Loam/sandy loam	13	60	$y = 1 - 114e^{0.132x}$	0.003	0.2
Sandy loam and sand						
Z3A	Sandy loam	13	44	$y = 1 + 126e^{-0.144x}$	0.002	19
X11A	Sand	13	190	$y = 2 + 176e^{-0.204x}$	0.002	47
Peat						
CU711A	Peat	10	98	$y = 3 + 45e^{-0.0746x}$	0.001	9
X18B	Peat	8	83	$y = 2 - 57e^{0.066x}$	0.031	0.4
X7A	Peat	9	97	$y = 9 - 191e^{0.220x}$	0.050	0.1

[a] Except 1st + 2nd cut in 1986 and 1st cut in 1987.
[b] Only fields with $p < 0.05$ are shown.

3.5. ^{137}Cs uptake and K and Ca content in grass

The potassium and calcium content in grass was analysed during the first six years following the initiation of the fieldwork (1987–1992). The potassium concentration in grass varied from 8.5 to 30.9 mg g^{-1} and calcium concentration varied from 3.6 to 10.3 mg g^{-1}. The transfer factors did not correlate with the concentration of potassium or calcium in grass tissue. However, the data reveal an insignificant negative correlation ($r = -0.53$) between the ^{137}Cs activity concentration and the potassium concentration in the plants grown on X fields with a high ^{137}Cs deposition level (100–200 kBq m^{-2}), while no such correlation was found for calcium (Fig. 3).

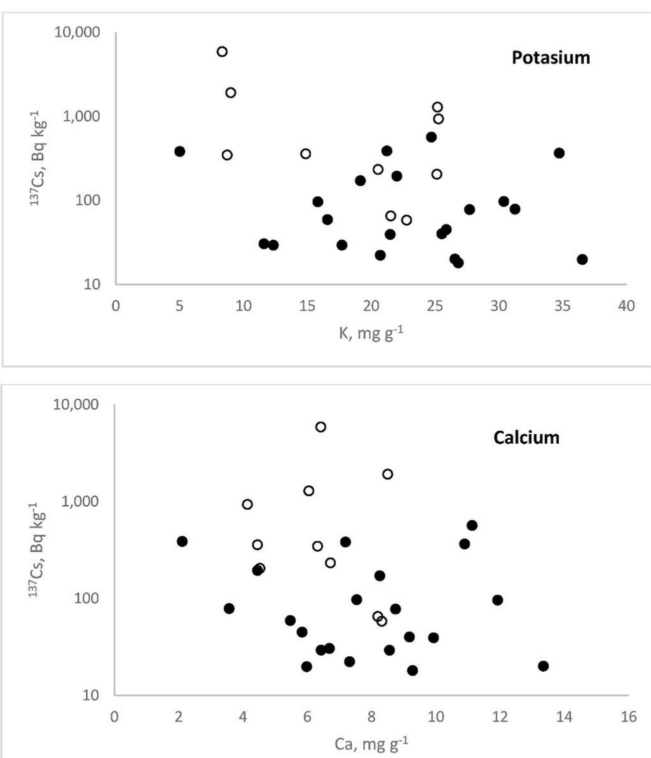

Fig. 3. Relationship of ^{137}Cs activity concentration (Bq kg^{-1}) vs potassium and calcium concentration in grass, mg g^{-1}, during the period 1987–1992 across all soil types, $n = 94$. Empty circles – fields with ^{137}Cs deposition 30–60 kBq m^{-2}; Filled circles – fields with ^{137}Cs deposition 100–200 kBq m^{-2}. No significant correlation either for potassium or for calcium, $p > 0.05$.

3.6. ^{137}Cs uptake and farming practices

The levels of ^{137}Cs transfer factors in grass on fields under intensive farming practices were generally lower compared to those in fields with extensive and moderate farming practices across all studied soil types, regardless radionuclide deposition density. The correlation between these two parameters was found to be weak ($r = -0.37$, $P = 0.07$).

3.7. Ecological half-life of ^{137}Cs

The ecological half-life of ^{137}Cs in grass is a result of all processes that contribute to the decrease in ^{137}Cs activity concentration over time. The median value was found to be 4.5 years, with a wide range of 2.3–18.4 years, and only partially depended on soil type (Fig. 4).

Half-lives exceeding eight years were absent in fields with more than 40% clay and fine silt, but appeared in approximately 25% of the

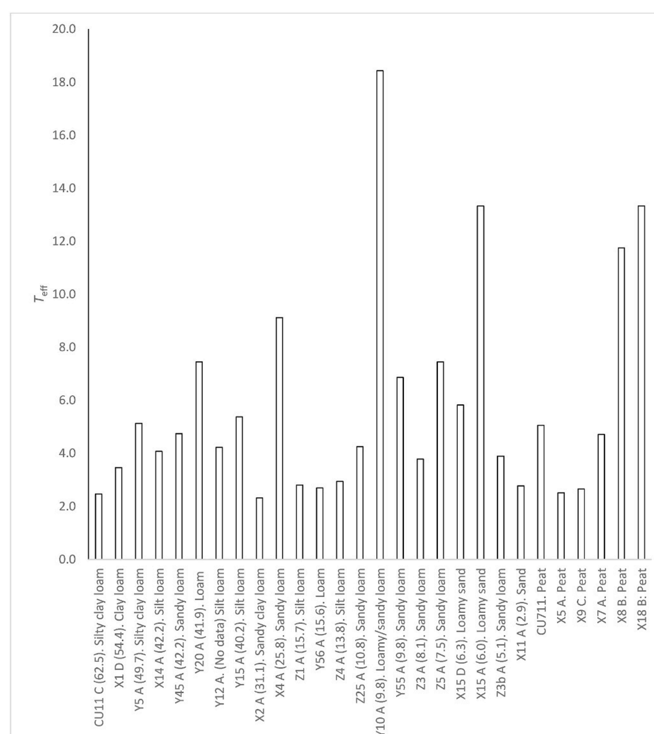

Fig. 4. Ecological half-life (T_{eff}, years) of ^{137}Cs in grass at different fields. The classification numbers of counties/fields: CU – Uppsala; X – Gävleborg; Y – Västernorrland; Z – Jämtland; figures and letters denote field numbers. The fields are ordered by their clay and fine silt content (in brackets, %).

remaining fields. The longest half-lives were observed on a peat soil (X18B) and on loam and sandy-loam soils with low clay content (7.0%, Y10A). However, short half-lives of 2–4 years occurred in fields on all types of soils. It was not significantly correlated with the investigated soil properties, including pH (H_2O), organic matter content, and texture.

4. Discussion

The ^{137}Cs transfer factors from soil to crops encompasses many processes including root uptake (IAEA, 2014). Caesium is taken up by plants in competition with potassium. Hence, the ^{137}Cs activity in harvested crops is controlled by the availability of both elements, which are both under the control by adsorption to clay minerals and organic matter, where particularly the former possess sites capable of relatively strong binding and may incorporate both caesium and potassium into a pool of ions that are fixed between collapsed 2:1 layers, with a reduced ability to become released by desorption (de Koning and Comans, 2004).

In our data, the importance of potassium availability was evident from the negative correlation between potassium concentration in grass and ^{137}Cs activity concentration in their tissues, which is in accordance with previous studies (Jones et al., 1991; Stone and Robison, 2002; Guillaume et al., 2012). This relationship was more pronounced in later years of the study.

Potassium availability in a given soil increases with an increase in exchangeable potassium, since less energetic binding sites become engaged in the binding of potassium with increasing coverage with the ion (Sparks and Huang, 1985). Under experimental conditions, the same holds for caesium ions. Although the absolute amounts of sorption increases when more caesium is added to a soil, there is a decreased relative tendency of binding, in terms of the distribution coefficient (Staunton, 1994). However, the mechanisms of caesium availability in the field are more complex, as illustrated by the fact that transfer factors were not correlated with the initial density of radionuclide deposition.

The deposition density of ^{137}Cs had a strong and direct impact on the absolute activity concentration of ^{137}Cs in grass shortly after the accident, but it had no significant effect on the transfer factor in a long-term perspective. This is in agreement with previous studies (Bunzl et al., 2000; Ciuffo et al., 2003). Instead, the transfer factor can be expected to vary with the abundance of clay minerals, in our study, estimated by the clay and fine-silt content (<0.006 mm). Our results suggest that rapid fixation of ^{137}Cs in silty clay loam and loam soils, particularly in upper soil layers, caused a drastic decrease in transfer factors already during the first few years after 1986, due to the well-known radionuclide association with clay minerals in the soil (Shenber and Eriksson, 1993; Delvaux et al., 2001). ^{137}Cs fixation is generally less effective in organic (peat) and sandy soils, where the vertical migration speed is generally higher (Rosén et al., 1999). However, the correlation between transfer factor and the clay and fine-silt content of the soil only accounted for a small portion, up to 18%, of the total variability, and Fig. 2 shows that there was considerable variation between our fields. Similarly, Nisbet and Woodman (2000) found that transfer factors for ^{137}Cs cannot be reliably predicted based on soil characteristics when analysing a large dataset.

The nanomolar, or less, concentration of $^{137}Cs^{+}$ added to soils as a consequence of the Chornobyl accident (Hird et al., 1996) was too low to induce, in itself, fixation due to interlayer collapse in clay minerals (de Koning and Comans, 2004). Fixation was more likely to occur because the coverage with K^+, or NH_4^+, exceeded a critical value (Sparks and Huang, 1985). It could be suspected, then, that soils with ample exchangeable potassium were prone to retain ^{137}Cs more firmly. However, our data on ^{137}Cs transfer to crops do not show any simple relationship with exchangeable potassium. A similar lack of a rational pattern was observed after forming different ratios of exchangeable potassium and clay content etc. This suggests that ^{137}Cs availability was controlled by temporal changes in potassium status that were not captured by the measurements of exchangeable potassium performed on single occasions.

By contrast, soil pH had a negative correlation (r = −0.54) with transfer factors, explaining approximately 30% of the total variability. It is well known that fixation of potassium in clay minerals is hampered by a low pH in the soil, due to competition with H_3O^+ ions on exchange sites and to formation of Al-hydroxy polycations that inhibit 2:1-layer collapse and thereby fixation by clay minerals (Sparks and Huang, 1985). Similar effects can be expected on the fixation of Cs^+ ions. Our results are in agreement with the findings of Schuller et al. (1988), who found that approximately 67% of the variance of transfer factors was explained by soil pH, while other soil parameters contributed only a few percent to the variance.

According to our data, transfer factors generally declined more slowly in the peat soils (Fig. 2e). However, there was considerable variation in the final levels across the fields. The transfer factors for the three fields on peat soil did not fall below 1×10^{-3} m^2 kg^{-1} throughout the study period. On two fields, this threshold was achieved within a span of 3–13 years, whereas one field even exhibited an increase over time. Shand et al. (1994) found that clay minerals may be present even in organic soils in sufficient quantity to fix caesium to some extent. The presence of these minerals would need further study to understand caesium dynamics in this category of soils. In mineral soils, presence of organic matter does inhibit Cs fixation by blocking sorption sites for Cs on clay minerals although the effect may be only moderate (Dumat et al., 1997; Dumat and Staunton, 1999). In our data, soil organic matter had no significant effect on the transfer factor. The continued increase in transfer factors in some fields on peat soils over time may be attributed to the gradual mineralization of plant residues contaminated by ^{137}Cs, which can take years (Moore et al., 2007), and to the low capacity of organic matter to retain ^{137}Cs (Koarashi et al., 2023). On the other hand, intensive farming practices, including soil ploughing, apparently reduced the ^{137}Cs transfer factor in one of the fields on peat soil (Fig. 2e).

The ecological half-life of ^{137}Cs is a crucial parameter for developing risk assessment models. Our estimated ecological half-life of 4.5 years for grass over all fields in the study is considerably shorter than the physical half-life of 30 years for ^{137}Cs, and is consistent with previous findings by Pröhl et al. (2006). The half-life varied widely and was not significantly influenced by soil type. The longest half-lives occurred in sandy and peaty soils, whereas the more clayey soils appeared more homogeneous. However, both long and short half-lives are possible on sandy and peaty soils.

5. Conclusions

After the Chornobyl accident, the transfer of ^{137}Cs from soil to grass decreased relatively rapidly in silty clay and loam soils, reaching a plateau below 1×10^{-3} m^2 kg^{-1} within 3–4 years and remaining stable for about 20 years. In contrast, peat, sandy loam, and loamy sand soils exhibited a slower decline of ^{137}Cs levels in the grass, resulting in median transfer factor values approximately 2–3 times higher on these fields compared to the average across all studied soils.

Our study suggests that several factors influence the transfer of ^{137}Cs from soil to grass. Specifically, we found that pH and clay content in soil are significant determinants of transfer factors, with pH accounting for up to 44% and clay content accounting for up to 18% of the total variability.

In contrast, no relationship was observed between transfer factors and exchangeable potassium in the soil. Potassium concentration in grass was not significantly and negatively correlated with ^{137}Cs activity concentration.

In silty clays and loams, the transfer factors decreased to a plateau below 1×10^{-3} m^2 kg^{-1} approximately 3–5 years following the fallout, whereas in soils with intermediate clay and silt content this process takes about 12–15 years. In most of peat soils, the transfer factors remained above 1×10^{-3} m^2 kg^{-1} even at the end of the study, that is, 21 years after the incident.

Growing grass on fields with high deposition density may be safe on soils with high clay content, as ^{137}Cs transfer from soil to grass significantly decreases after the year when the fallout occurred.

CRediT authorship contribution statement

Mykhailo Vinichuk: Writing – review & editing, Writing – original draft, Visualization, Conceptualization. **Magnus Simonsson:** Writing – review & editing, Data curation. **Maja Larsson:** Investigation, Data curation. **Klas Rosén:** Writing – review & editing, Investigation, Data curation, Conceptualization.

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.

Acknowledgements

The principal author received a grant from the Swedish University of Agricultural Sciences (SLU) and the Royal Swedish Academy of Sciences.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2025.107621.

Data availability

Data will be made available on request.

References

Brimo, K., Gonze, M.A., Pourcelot, L., 2020. Long term decrease of ^{137}Cs bioavailability in French pastures: results from 25 years of monitoring. J. Environ. Radioact. 208–209, 106029. HAL Id: hal-02454921. https://hal.science/hal-02454921.

Bunzl, K., Albers, B.P., Schimmack, W., Belli, M., Ciuffo, L., Menegon, S., 2000. Examination of a relationship between Cs-137 concentrations in soils and plants from alpine pastures. J. Environ. Radioact. 48, 145–158. https://doi.org/10.1016/S0265-931X(99)00061-2.

Ciuffo, L., Velasco, H., Belli, M., Sansone, U., 2003. Cs-137 soil-to-plant transfer for individual species in a semi-natural grassland. Influence of potassium soil content. J. Radiat. Res. 44, 277–283. https://doi.org/10.1269/jrr.44.277.

Coughtrey, P.J., Thorne, M.C., 1983. Radionuclide Distribution and Transport in Terrestrial and Aquatic Ecosystems: a Critical Review of Data, vol. 1. A. A. Balkema, Rotterdam, pp. 330–337.

Day, P.R., 1965. Hydrometer method of particle size analysis. In: Black, C.A. (Ed.), Methods of Soil Analysis. American Society of Agronomy, Madison, Wisconsin Argon, pp. 562–563. https://doi.org/10.2134/agronmonogr9.1.c43.

de Koning, A., Comans, R.N.J., 2004. Reversibility of radiocaesium sorption on illite. Geochem. Cosmochim. Acta 68, 2815–2823. https://doi.org/10.1016/j.gca.2003.12.025.

Delvaux, B., Kruyts, N., Maes, E., Smolders, E., 2001. Fate of radiocesium in soil and rhizosphere. In: Gobran, G.R., Wenzel, W.W., Lombi, E. (Eds.), Trace Elements in the Rhizosphere. CRC Press, London, pp. 61–91.

Dumat, C., Staunton, S., 1999. Reduced adsorption of caesium on clay minerals caused by various humic substances. J. Environ. Radioact. 46, 187–200. https://doi.org/10.1016/S0265-931X(98)00125-8.

Dumat, C., Cheshire, M.V., Fraser, A.R., Shand, C.A., Staunton, S., 1997. The effect of removal of soil organic matter and iron on the adsorption of radiocaesium. Eur. J. Soil Sci. 48, 675–683. https://doi.org/10.1111/j.1365-2389.1997.tb00567.x.

Egnér, H., Riehm, H., Domingo, W.R., 1960. Untersuchungeniiber die chemische Bodenanalyse als Grundlage fiir die Beurteilung des Nahrstoffzustandes der Boden. II. Chemische Extraktionsmethoden zur Phosphor und Kaliumbestimmung. K Lantbr högsk Alnarp (Uppsala) 26, 199–215, 1960.

Guillaume, T., Chawla, F., Steinmann, P., Gobat, J.-M., Froidevaux, P., 2012. Disparity in ^{90}Sr and ^{137}Cs uptake in Alpine plants: phylogenetic effect and Ca and K availability. Plant Soil 355 (1–2), 29–39. https://doi.org/10.1007/s11104-011-1110-6.

Hird, A.B., Rimmer, D.L., Livens, F.R., 1996. Factors affecting the sorption and fixation of caesium in acid organic soil. Eur. J. Soil Sci. 47, 97–104. https://doi.org/10.1111/j.1365-2389.1996.tb01376.x.

IAEA, 2014. Handbook of Parameter Values for the Prediction of Radionuclide Transfer to Wildlife. IAEA, Vienna. Technical Reports Series no. 479.

Isaksson, M., Erlandsson, B., Linderson, M.-L., 2000. Calculation of the deposition of ^{137}Cs from the nuclear bomb tests and from the Chernobyl accident over southern Sweden based on the precipitation. J. Environ. Radioact. 49, 97–112. https://doi.org/10.1016/S0265-931X(99)00101-0.

Jones, H., Harrison, A.F., Poskitt, J.M., Roberts, J.D., Clint, G., 1991. The effect of potassium nutrition on ^{137}Cs uptake in two Upland species. J. Environ. Radioact. 14, 279–294. https://doi.org/10.1016/0265-931X(91)90019-C.

Koarashi, J., Atarashi-Andoh, M., Nishimura, S., 2023. Effect of soil organic matter on the fate of ^{137}Cs vertical distribution in forest soils. Ecotoxicol. Environ. Safety 262, 115177. https://doi.org/10.1016/j.ecoenv.2023.115177.

Krouglov, S.V., Filipas, A.S., Alexakhin, R.M., Arkhipov, N.M., 1997. Long-term study on the transfer of ^{137}Cs and ^{90}Sr from Chernobyl-contaminated soils to grain crops. J. Environ. Radioact. 34 (3), 267–286. https://doi.org/10.1016/0265-931X(96)00043-4.

Moore, T.R., Bubier, J.L., Bledzki, L., 2007. Litter decomposition in temperate peatland ecosystems: the effect of substrate and site. Ecosystems 10, 949–963, 2007.585 2007. https://www.jstor.org/stable/27823735.

Ng, Y.C., 1982. A review of transfer factors for assessing the dose from radionuclides in agricultural products. Nucl. Saf. 23 (1), 57–71.

Nisbet, A.F., Woodman, R.F.M., 2000. Soil-to-plant transfer factors for radiocaesium and strontium in agricultural systems. Health Phys. 78 (3), 279–288. https://doi.org/10.1097/00004032-200003000-00005.

Pröhl, G., 2009. Interception of dry and wet deposited radionuclides by vegetation. J. Environ. Radioact. 100, 675–682. https://doi.org/10.1016/j.jenvrad.2008.10.006.

Pröhl, G., Ehlken, S., Fiedler, I., Kirchner, G., Klemt, E., Zibold, G., 2006. Ecological half-lives of Sr and Cs in terrestrial and aquatic ecosystems. J. Environ. Radioact. 91 (1–2), 41–72. https://doi.org/10.1016/j.jenvrad.2006.08.004.

Rosén, K., 1996. Transfer of radiocaesium in sensitive agricultural environments after the Chernobyl fallout in Sweden. II. Marginal and seminatural areas in the county of Jämtland. Sci. Total Environ. 182 (1−3), 135–145. https://doi.org/10.1016/0048-9697(95)05059-0.

Rosén, K., Eriksson, Å., Haak, E., 1996. Transfer of radiocaesium in sensitive agricultural environments after the Chernobyl fallout in Sweden. I. County of Gavleborg. Sci. Total Environ. 182 (1−3), 117–133. https://doi.org/10.1016/0048-9697(95)05056-6.

Rosén, K., Eriksson, Å., Haak, E., 1998. Transfer of radiocaesium in sensitive agricultural environments after the Chernobyl fallout in Sweden: III. County of Västernorrland. Sci. Total Environ. 209 (2−3), 91–105. https://doi.org/10.1016/S0048-9697(98)80100-9.

Rosén, K., Öborn, I., Lönsjö, H., 1999. Migration of radiocaesium in Swedish soil profiles after the Chernobyl accident, 1987–1995. J. Environ. Radioact. 46 (1), 45–66. https://doi.org/10.1016/S0265-931X(99)00040-5.

Sarap, N.B., Janković, M.M., Dolijanović, Z.K., Kovačević, D.D., Rajačić, M.M., Dragana, J.D., Todorovic, J., 2015. Soil-to-plant transfer factor for ^{90}Sr and ^{137}Cs. J. Radioanal. Nucl. Chem. 303, 2523–2527. 10.1007/s10967-014-3809-3.

Schuller, P., Handl, J., Trumper, R.E., 1988. Dependence of the ^{137}Cs soil to plant transfer factor on soil parameters. Health Phys. 55 (3), 575–577.

SGAB, 1986. Swedish Geological Company, Map of ^{137}Cs kBq m^2, Ground surface results from aerial surveys, May to October 1986. Uppsala. https://www.sgu.se/om-sgu/verksamhet/kartlaggning/geofysik_att_se_ner_i_berget/flyggeofysisk-matning/flygmatningar-efter-tjernobyl/.

Shand, C.A., Cheshire, M.V., Smith, S., Vidal, M., Rauret, G., 1994. Distribution of radiocesium in organic soils. J. Environ. Radioact. 23, 285–302. https://doi.org/10.1016/0265-931X(94)90067-1.

Shenber, M.A., Eriksson, A., 1993. Sorption behaviour of caesium in various soils. J. Environ. Radioact. 19 (1), 41–51. https://doi.org/10.1016/0265-931X(93)90057-E.

Smith, J.T., Fesenko, S.V., Howard, B.J., Horril, A.D., Sanzharova, N.I., Alexakhin, R.M., Elder, D.G., Naylor, C., 1999. Temporal change in fallout ^{137}Cs in terrestrial and aquatic systems: a whole ecosystem approach. Environ. Sci. Technol. 33, 49–54. https://doi.org/10.1021/es980670t.

Sparks, D.L., Huang, P.M., 1985. Physical chemistry of soil potassium. In: Munson, R.D. (Ed.), Potassium in Agriculture. Soil Science Society of America, Madinson, Wisconsin, pp. 201–276. https://doi.org/10.2134/1985.potassium.c9.

Staunton, S., 1994. Adsorption of radiocaesium on various soils: interpretation and consequences of the effects of soil: solution ratio and solution composition on the distribution coefficient. Eur. J. Soil Sci. 45, 409–418. https://doi.org/10.1111/j.1365-2389.1994.tb00526.x.

Stone, E.L., Robison, W.L., 2002. Effect of potassium on uptake of ^{137}Cs in food crops grown on coral soils: annual crops at bikini atoll. Experiment 4. Effects of Stable 133Cs and K Additions on ^{137}Cs in Sorghum. Lawrence Livermore National Laboratory. Technical Information Department's Digital Library. http://www.llnl.gov/tid/Library.html.

Tsukada, H., Hasegawa, H., 2002. Soil-to-plant transfer of ^{137}Cs and other essential and trace elements in cabbage plants. J. Radioanal. Nucl. Chem. 252 (2), 219–224. 10.1023/A:1015789500124.

Uchida, S., Tagami, K., 2007. Soil-to-plant transfer factors of fallout ^{137}Cs and native ^{133}Cs in various crops collected in Japan. J. Radioanal. Nucl. Chem. 273, 1. https://doi.org/10.1007/s10967-007-0737-5.

Yasuda, H., 1995. Transfer models in a soil-plant system used for environmental impact assessments. J. Nucl. Sci. Technol. (Tokyo, Jpn.) 32 (12), 1272–1283. https://doi.org/10.1080/18811248.1995.9731850.

Yoshikawa, S., Igura, M., Yasutaka, T., Eguchi, S., 2020. Physicochemical and time factors affecting ^{137}Cs transfer through a paddy soil–rice system. Soil Sci. Plant Nutr. 66 (4), 541–552. https://doi.org/10.1080/00380768.2020.1787785.