Chemosphere 366 (2024) 143525

Contents lists available at ScienceDirect

Chemosphere

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

Long-term effects of potassium chloride, wood ash, and EDTA on ^{137}Cs soil-plant transfer in a mixed forest of Ukraine

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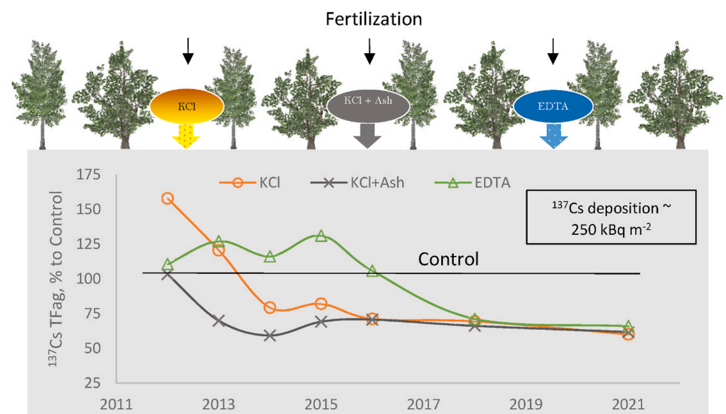
HIGHLIGHTS

- Forest soil was fertilised to counteract ^{137}Cs uptake by shrub and tree species.
- ^{137}Cs uptake reduction across species due to KCl application was inconsistent.
- KCl + wood ash substantially decrease ^{137}Cs uptake by forest plants.
- EDTA had no statistically significant effect on ^{137}Cs uptake by studied plants.

GRAPHICAL ABSTRACT



Average ^{137}Cs TFag decrease in forest vegetation after fertilization with KCl, KCl+Ash and EDTA

ARTICLE INFO

Handling editor: Milena Horvat

Keywords:
Fertilisation
Forest soil
Wood ash
Potassium
Zinc
Manganese

ABSTRACT

We conducted a 10-year field experiment to study the effects of potassium chloride, wood ash, zinc, and manganese on reducing ^{137}Cs uptake by young leaves and green shoots of common dwarf shrubs and tree species near the Chornobyl Nuclear Power Plant. A field experiment had four treatments: a control with no fertilisation, and three fertilised treatments: potassium fertiliser (KCl), a combination of potassium fertiliser and wood ash (KCl + Ash), and a solution providing zinc and manganese (EDTA). There was approximately 30 % decrease in ^{137}Cs uptake by most of the studied plants species growing on plots fertilised with KCl compared to unfertilised plots during intermediate (2014–2016) and late (2018–2021) periods. Combining KCl with wood ash was found to be the most effective countermeasure, reducing ^{137}Cs uptake by up to 60% in most species, while treatment with EDTA was less effective. Generally, the decline in ^{137}Cs uptake by plants over the study years following treatments with fertilisers was more pronounced than in the control, indicating the efficiency of fertilisation in

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https://doi.org/10.1016/j.chemosphere.2024.143525
Received 19 June 2024; Received in revised form 20 September 2024; Accepted 10 October 2024
Available online 10 October 2024

reducing ^{137}Cs uptake by forest plants. Our research suggests that a combination of potassium chloride and wood ash can still effectively reduce ^{137}Cs transfer in most common forest species, even years after the accident.

1. Introduction

Caesium-137 (^{137}Cs) was one of the major fission products released in the accident at Chornobyl Nuclear Power Plant (CNPP) in 1986 and at Fukushima Nuclear Power Plant in 2011. This radionuclide has a physical half-life of around 30 years and is still present in high concentrations in contaminated soils (Vinichuk et al., 2023). Forest ecosystems are known to act as a filter, intercepting, distributing, and retaining radionuclides (Kiefer et al., 1996). Even many years after the CNNP accident, ^{137}Cs is still mostly concentrated in the upper 0–15 cm soil layer in contaminated natural and semi-natural environments (Vinichuk and Johanson, 2003; Lehto et al., 2013), and is thus available for plant uptake. The uptake of ^{137}Cs by forest plants, animals and fungi was very high in the first few years after the accident and has decreased slowly over time (Palo et al., 2003; Strzałek et al., 2021).

Countermeasures such as soil fertilisation and liming have been used successfully to reduce radionuclide transfer from soil to crop plants in agricultural ecosystems (Rosén and Vinichuk, 2014; Rosén et al., 2011). While these countermeasures have been effective in agricultural ecosystems, they have received less attention in the context of forest ecosystems. Only a few suitable countermeasures have been developed for forest soils (Levula et al., 2000; Zibold et al., 2009; Aro and Rantavaara, 2011)

A long-lasting inhibiting effect of potassium (K) fertilisation on ^{137}Cs uptake by pine trees, growing on peat soil has been reported in Finland (Kaunisto et al., 2002). A similar effect of a single K application (100 kg K ha^{-1}) to forest soils on ^{137}Cs uptake by green aboveground parts of heather, lingonberry, bilberry and several fungal species has been observed in Swedish forests (Rosén et al., 2014). Suppressive effects of K fertilisation on ^{137}Cs uptake by hinoki cypress seedlings have also been reported following the Fukushima accident in Japan (Komatsu et al., 2017).

Woodlands in Ukraine with ^{137}Cs surface contamination exceeding 37 kBq m^{-2} occupy a total area of about 13,000 km^2 (IAEA, 2001). To our knowledge, no previous study has examined the effects of fertilisation on ^{137}Cs uptake by vegetation in these highly contaminated forests. To address this knowledge gap, we investigated ^{137}Cs uptake by young leaves and green shoots of small woody, evergreen dwarf shrubs and common tree species in the forest ecosystem of Ukraine after a single application of potassium chloride (KCl) fertiliser, combined KCl fertiliser and wood ash or chelated ethylene diamine tetra acetic acid (EDTA) containing the micronutrients zinc (Zn) and manganese (Mn). Both mineral nutrients in the form of commercial fertiliser or wood ash were previously tested and recommended for the remediation of forests contaminated with ^{137}Cs (Levula et al., 2000; Aro and Rantavaara, 2011). However, the combined effect of KCl fertiliser and wood ash treatment has not been studied yet. We also included EDTA as a treatment since sandy soils in north-western Ukrainian forest ecosystems are generally Zn and Mn deficient (Polupan, 1988). Chelated EDTA was used to provide plants with available Zn and Mn and to examine the effect of these microelements on ^{137}Cs uptake by forest plants.

We hypothesized that fertilisation of forest soil can reduce ^{137}Cs uptake by common shrub and tree species, mainly due to higher soil pH and increased concentration of K^+ ions in the soil solution. Specific objectives of the study were to: (1) investigate the effect of a single application of KCl, KCl combined with wood ash, and EDTA on ^{137}Cs uptake by young leaves and annual shoots of common species of trees and dwarf shrubs; (2) evaluate the long-term response of ^{137}Cs uptake by different plant species to a one-time application of selected fertilisers.

2. Material and methods

2.1. Site description

Our field study site was established in a mixed forest, near Bazar village, within the Drevlianskyi Nature Reserve (51.05°35′N, 29.18°56′E), situated approximately 70 km from the Chornobyl Nuclear Power Plant (CNPP) in Zhytomyr region, north-western Ukraine (Fig. 1). During 2012–2014, the deposition of ^{137}Cs in this area ranged between 177 and 355 kBq m^{-2} (Table S1).

The soil at the site is a soddy podzolic soil, comprising 92.8% ± 1.53% sand (with a high content of silica), 6.9% ± 1.53% silt and clay, developed on glacial-fluvial sediment. The thickness of the organic horizon generally does not exceed 3–5 cm. The main chemical characteristics of the topsoil (0–15 cm layer) at the site before the start of the experiment are presented in Table 1.

The dominant tree species in the area are Scots pine (*Pinus sylvestris*), with some intermixed birch (*Betula pendula*), rowan (*Sorbus aucuparia*), oak (*Quercus robur*), and alder buckthorn (*Frangula alnus*). The most common dwarf shrubs are bilberry (*Vaccinium myrtillus*), lingonberry (*Vaccinium vitis-idaea*), bracken (*Pteridium aquilinum*) and heather (*Calluna vulgaris*). The forest floor and tree bark are partly covered by various mosses and lichens, mainly *Dicranum polysetum* and *Dicranum scoparium*. The stand was approximately 60–80 years old at the beginning of the experiment.

2.2. Experiment layout

The field experiment was conducted between 2012 and 2021 and consisted of four treatments (Fig. 1): 1) No fertilization (control); 2) KCl fertilisers, supplying 100 kg K ha^{-1}; 3) KCl fertilisers in combination with wood ash, supplying 100 kg K ha^{-1} (half from KCl and half from wood ash), denoted as treatment KCl + Ash; and 4) micronutrients applied in the form of EDTA solution in the amount recommended by the manufacturer, i.e. 0.5–1.0 L ha^{-1}, supplying 25% of Zn and 20% of Mn, dissolved in 80 L of water, denoted as treatment EDTA. The wood ash used had a pH of about 12.8, a total potassium content of 3.1%, a total phosphorus content of 0.9%, and an activity concentration (AC) of ^{137}Cs of 46 Bq kg^{-1}. All fertilizers were applied manually to the forest floor on one occasion in April 2012. Each treatment had four replicates, resulting in a total of 16 plots (each 20 m × 10 m) situated within a 0.32 ha area of the forest (Fig. 1). Treatments were randomly assigned to these 16 experimental plots.

2.3. Sampling

Two soil samples were taken from each of the 16 plots before fertiliser application in 2012 and two years after application, in 2014. The soil was sampled to a depth of 15 cm using cylindrical steel cores with a diameter of 57 mm and a height of 150 mm. All soil samples were air-dried at a maximum temperature of 30 °C for approximately two weeks, then milled, sieved through a 2-mm mesh to produce homogeneous material, and weighed for radiometric and chemical analyses. For chemical analysis, 10 g samples of air-dried, milled, and sieved soil collected in 2012 were used.

Post-treatment samples of aboveground biomass from dwarf shrubs and young shoots of trees were collected randomly from each plot every month from April to September in 2012. Prior to fertilisation in April, no aboveground biomass was available for sampling, except of one sample of bilberry and cowberry plants from each treatment. In subsequent years (2013, 2014, 2015, 2016, 2018, and 2021), samples were collected from each plot one to three times per growing season (late June–early July). A total of approximately 1340 (16 plots x 7 species x 12 samplings) plant samples were air-dried, cut into smaller pieces, thoroughly mixed, weighed, and placed in standard 35- or 60-mL plastic tubes for gamma spectrometry measurements. Additionally, 16 soil samples were randomly collected prior to the start of the experiment in April 2012, and 16 samples were collected two years later in 2014. Bilberry berries were collected occasionally when available, dried at 65 °C, weighed, and analysed for ^{137}Cs AC.

2.4. Radiometry and data processing

Activity concentration of ^{137}Cs (Bq kg^{-1} dry weight (d.w.)) in plant and soil samples was measured using HPGe and NaI detectors for no longer than 24 h, to achieve an error of less than 5%. All results were decay-corrected to the sampling date. Data were processed using WinDAS™ (Windows Data Acquisition System) and Apex-Gamma™ software.

Soil contamination density (A_s, Bq m^{-2}) was calculated as:

$$A_s = \frac{A_m \times m}{S} \tag{1}$$

where A_m is the activity concentration of ^{137}Cs per unit dry weight of soil on the sampling date (Bq kg^{-1}), m is weight of soil in the cylindrical steel core (kg) and S is area of the core (m^2).

Aggregated transfer factor (TF_{ag}, m^2 kg^{-1}), i.e. the coefficient of ^{137}Cs transfer from soil to plant material, was then calculated as:

$$TF_{ag} = \frac{A_m}{A_s} \tag{2}$$

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 average level of soil contamination with ^{137}Cs (Bq m^{-2}) recalculated for the sampling date.

The rate of decrease in ^{137}Cs TF_{ag} over the study period for the different plant species was calculated as a start/end ratio, i.e. TF_{ag} in 2012 divided by TF_{ag} in 2021.

2.5. Statistical analysis

The effect of the fertiliser treatments on the different shrub and tree species over the 10-year study period was tested using a repeated measure mixed effect model. The sampling plot was treated as a random factor ($n = 56$) and plot treatment (fertilised or control), log-transformed ^{137}Cs AC in plant samples, year, and interaction between

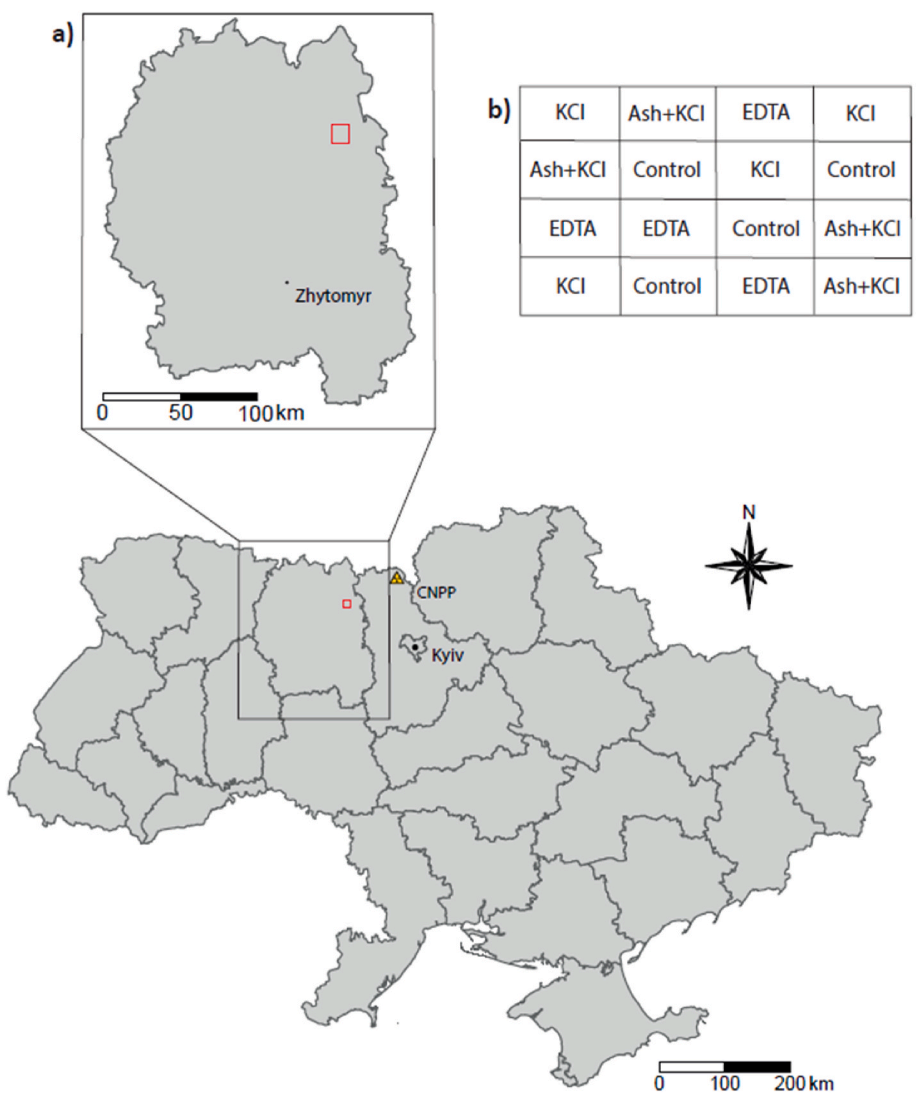

Fig. 1. Experimental design: **a**) study area in Bazar forest, north-western Ukraine, and **b**) layout of treatment plots. Control = non-fertilised plots; KCl = potassium fertiliser; KCl + Ash = potassium fertiliser combined with wood ash; EDTA = solution supplying Zn and Mn.

plot treatment and year was included as candidate explanatory variables. The model was fitted using the R-package *nlme* (Pinheiro et al., 2022).

To evaluate the difference between the treatments and control an analysis of variance (ANOVA) was performed using R (R Core Team, 2020) and Minitab® 9.2020.1 statistical software.

3. Results

The mean ^{137}Cs deposition within the study area was 250 kBq m^{-2} (range 239–287 kBq m^{-2}) (Table S1). The deposition levels recorded in individual cores in the study area were still relatively high (up to 480 kBq m^{-2}).

3.1. ^{137}Cs activity concentration

The treatment effect on ^{137}Cs AC in plants and ^{137}Cs TF$_{ag}$ from soil to plants was compared over three periods: early (2012–2013), intermediate (2014–2016), and late (2018–2021). ^{137}Cs AC in young leaves and green shoots of the studied plant species varied between 0.24 and 8.70 kBq kg^{-1}, with the highest values recorded at the beginning of the experiment (2012) and the lowest at the end (2021). The lowest ^{137}Cs AC was found in young leaves and green shoots of alder buckthorn (0.24–1.43 kBq kg^{-1}) and the highest in oak leaves and shoots (0.75–7.24 kBq kg^{-1}) (Table S2). The mean ^{137}Cs AC in treated plots was often higher than in control plots during the early years (2012–2013), but lower in the later years (2018–2021).

3.2. Aggregated transfer factor

Overall, the ^{137}Cs TF$_{ag}$ from soil to young leaves and green shoots decreased in all studied tree and dwarf shrub species throughout the study period (Table S3). The rate of decrease was about twice as fast for species growing in fertilised plots compared to non-fertilised plots. During the first year, the TF$_{ag}$ for young shoots and leaves of dwarf shrubs and trees in control plots ranged from 5.4 to 13.5 $\times$ 10^{-3} m^2 kg^{-1} but decreased to 1.7–7.4 $\times$ 10^{-3} m^2 kg^{-1} by 2021 (Table 2). In fertilised plots, the TF$_{ag}$ decreased by an average 3–5-fold (Table 2). Over the study period, the highest significant ($p < 0.01$) decrease in mean TF$_{ag}$ was observed for plants growing in K-fertilised plots (KCl, KCl + Ash), with a less pronounced, yet still significant decrease for certain species in control plots (Table 2).

3.3. Fertilisation with KCl

3.3.1. Bilberry

Uptake of ^{137}Cs by bilberry plants sampled prior to fertilisation on the KCl treatment plot appeared to be about four times higher than in the control (Fig. 2b). Even though only a single sample of bilberry plants showing high ^{137}Cs TF$_{ag}$ was available and analysed in April 2012, a similar pattern remained evident in the following months after fertilisation, when samples from four replicates were analysed. Consequently, ^{137}Cs uptake by bilberry plants on the KCl treatment was consistently higher compared to the control in the first, second, and even third year after application (Figs. 2b, 3b).

In the intermediate period after fertilisation (2014–2016), the transfer of ^{137}Cs from soil to young leaves and green shoots of bilberry was similar in KCl-fertilised and control plots, while on subsequent measuring occasions (2018–2021) the ^{137}Cs TF$_{ag}$ values in plants growing in fertilised plots were about 40 % lower than in plants growing in control plots (Fig. 3b). The ANOVA results indicated that ^{137}Cs TF$_{ag}$ for young shoots and leaves of bilberry in 2021, i.e. in the ninth year after the application, was not statistically significantly lower following fertilisation with KCl. Bilberry berries from KCl-fertilised plots had significantly higher ^{137}Cs TF$_{ag}$ values initially, especially in the first two years after K application (Fig. 3c), but after three years since fertilisation, ^{137}Cs uptake by berries was similar in fertilised and control plots. The decline ratios in ^{137}Cs TF$_{ag}$ for both bilberry plants and berries in the KCl plots during the experiment were significantly higher (6.3 and 11.1) than those in the control plots (1.7 and 2.0), indicating the high efficiency of potassium fertilisers (Table 2).

3.3.2. Lingonberry

Similarly to bilberry, the ^{137}Cs uptake by lingonberry plants on KCl treatment plots prior to fertilisation was about 3 times higher than in plants growing on control plots (Fig. 2e), which effectively eliminated the effect of KCl application. However, the difference in ^{137}Cs transfer from soil to young lingonberry shoots and leaves on KCl and control plots levelled out within the first growing season after fertilisation (April–September 2012) (Fig. 2e). In the following years, TF$_{ag}$ from soil to plants growing in KCl-fertilised plots were generally 20–30 % lower than for plants growing in control plots. Notably, the effect of K fertilisation on ^{137}Cs uptake by lingonberry plants was still not statistically significant (Fig. 3e), likely due to differences in uptake before the beginning of the experiment. The decline ratio in ^{137}Cs TF$_{ag}$ for lingonberry plants in the KCl plots during the experiment was twice as pronounced as that in the control plots (Table 2).

3.3.3. Rowan, alder buckthorn, birch and oak

Application of KCl fertiliser did not affect ^{137}Cs uptake by rowan in early (2012–2013) and intermediate (2014–2016) years after treatment, but reduced ^{137}Cs TF$_{ag}$ by about 30% in later (2018–2021) years. ^{137}Cs uptake by alder buckthorn did not change in the early years after potassium fertilisation, but decreased by the approximately 25 and 50 % in the intermediate and late periods ($P = 0.047$) respectively. Radionuclide uptake by leaves and shoots of birch trees doubled in the first and second years after fertilisation, followed by a decrease of about 50 % after 2016. ^{137}Cs TF$_{ag}$ for young leaves and green shoots of oak trees growing in KCl-fertilised plots were higher than in control plots during the first two years after fertilisation, but approximately 36 % lower during 2014–2016. ^{137}Cs TF$_{ag}$ was not significantly different from control plots during the late years (2018–2021).

Generally, the decline ratio of ^{137}Cs uptake for all tree species during the experiment following fertilisation with KCl was up to 3 times more pronounced than on the control (Table 2). The difference between the KCl and control plots at the beginning of the experiment may be attributed to factors other than fertilisation. Additionally, for all species except birch, the difference in TF$_{ag}$ between the KCl and control plots decreased during 2012, indicating a potential positive effect of KCl application. For birch, the effect was delayed until 2014 (Fig. 3d).

Table 1
Chemical characteristics of the topsoil (0–15 cm layer) at the study site. Mean ± SD, n = 14.

pH	Carbon total, %	Nitrogen	C/N ratio	K-AL[a]	Ca-AL[b]	K–HCl[c]	Ca–HCl[d]
				mg 100g^{-1}			
4.25 ± 0.47	2.96 ± 1.93	0.12 ± 0.09	24.76 ± 2.78	7.03 ± 6.36	16.82 ± 19.22	15.63 ± 6.31	22.85 ± 21.70

[a] Ammonium acetate-lactate extractable potassium.
[b] Ammonium acetate-lactate extractable calcium.
[c] Acid-extractable potassium.
[d] Acid-extractable calcium.

Table 2
Change in mean aggregated transfer factor (TF_{ag}) for ^{137}Cs in forest plants between sampling during 2012 and 2021 (n = 4). The decline ratio is calculated as the mean ^{137}Cs TF_{ag} in 2012 divided by the mean ^{137}Cs TF_{ag} in 2021. Significant correlations between the decline ratio and year (p > 0.05) are highlighted in bold.

Species	^{137}Cs TF_{ag} × 10^{-3} m^2 kg^{-1}		Decline ratio	p-value	^{137}Cs TF_{ag} × 10^{-3} m^2 kg^{-1}		Decline ratio	p-value	^{137}Cs TF_{ag} × 10^{-3} m^2 kg^{-1}		Decline ratio	p-value	^{137}Cs TF_{ag} × 10^{-3} m^2 kg^{-1}		Decline ratio	p-value
	2012	2021			2012	2021			2012	2021			2012	2021		
	Control				KCl				KCl + Ash				EDTA			
Alder buckthorn	5.42	2.80	1.9	**0.04**	5.95	1.23	4.8	**0.02**	5.49	2.02	2.7	0.18	2.47	2.18	1.2	0.19
Bilberry	12.44	7.25	1.7	**0.04**	20.31	3.20	6.3	**0.01**	10.45	2.17	4.8	**0.01**	16.22	2.94	5.5	**0.01**
Bilberry berries	10.02	5.07	2.0	0.09	33.18	2.99	11.1	0.06	15.75	2.36	6.7	0.09	11.49	3.83	3.0	**0.04**
Birch	7.24	6.80	1.1	0.23	15.57	3.30	4.7	**0.03**	8.51	3.48	2.4	**0.05**	6.79	5.36	1.3	0.42
Lingonberry	13.28	6.17	2.2	**0.01**	22.47	5.02	4.5	**0.05**	10.58	5.60	1.9	0.17	21.8	3.75	5.8	**0.01**
Oak	8.48	4.63	1.8	0.09	25.92	4.42	5.9	**0.04**	15.41	5.48	2.8	**0.02**	14.3	2.63	5.4	0.11
Rowan	13.46	7.43	1.8	0.13	11.65	4.51	2.6	0.06	7.03	3.09	2.3	0.08	9.18	5.59	1.6	0.22

3.4. Fertilisation with KCl-Ash

3.4.1. Bilberry

Fertilisation of soil with KCl + Ash reduced the ^{137}Cs TF_{ag} from soil to young leaves and green shoots of bilberry already during the first year after fertilisation, compared to bilberry plants growing in control plots (Fig. 2b).

The effect remained significant even after 5–9 years (Fig. 3b). In the period 2012–2013, the transfer of ^{137}Cs from soil to bilberry plants was around 30% lower for plants growing in KCl + Ash-fertilised plots than for those growing in control plots. This effect was even stronger in the intermediate (approximately 40 % reduction, P = 0.033) and late (64 % reduction, P = 0.040) periods. The TF_{ag} from soil to bilberry berries were higher in the first year after fertilisation (Figs. 2c) and 30–40 % lower in the following years on treatment plots compared to control (Fig. 3c), although the difference was not significant. The difference in the decline ratio of ^{137}Cs uptake for bilberry plants and berries was three times more pronounced in plants growing in KCl + Ash-fertilised compared to those in control plots (Table 2). This indicates that the combined application of potassium and wood ash for reducing of ^{137}Cs transfer from soil to plants is an effective countermeasure, even though it may take some time after fertilisation to become evident.

3.4.2. Lingonberry

Fertilisation with KCl-Ash did not change ^{137}Cs uptake by young leaves and green shoots of lingonberry plants in the first year after treatment (Fig. 3e). However, KCl-Ash reduced ^{137}Cs uptake significantly during the periods 2013–2014 (by approximately 38 %, p = 0.022), 2014–2016 (by approximately 46 %, P = 0.006) and 2018–2021 (by approximately 24 %) periods (Fig. 3e). The decline ratio of ^{137}Cs TF_{ag} with time for lingonberry plants on fertilised and unfertilised plots was about the same (Table 2).

3.4.3. Rowan, alder buckthorn, birch and oak

Young leaves and green shoots of rowan trees growing in KCl-Ash plots had significantly lower (44 %, P = 0.041) ^{137}Cs TF_{ag} compared to controls during the first two years after fertilisation. The positive effect of fertilisation was even stronger (about 50 % reduction, P = 0.006) in the following periods (2014–2021). Fertilisation of alder buckthorn trees with KCl-Ash had only a marginal (20–25 %) and non-significant effect on the reduction of ^{137}Cs uptake from soil. Overall, uptake of ^{137}Cs by young leaves and green shoots of oak trees was stable throughout the study (Figs. 2f, 3f), while ^{137}Cs transfer from soil to leaves and shoots of birch trees reduced noticeably (about 60%) although not-significantly only in the late years (2018–2021) (Figs. 2d, 3d). The decline ratio of ^{137}Cs TF_{ag} over time for tree species on fertilised plots was generally 1.5 times higher than that for those growing on unfertilised plots (Table 2).

3.5. Fertilisation with EDTA

3.5.1. Bilberry and lingonberry

EDTA fertilisation (addition of Zn and Mn) did not reduce ^{137}Cs uptake by leaves, green shoots, and berries of bilberry in the early (2012–2013) and intermediate (2014–2016) periods, followed by 30–40 % lower uptake in the late (2018–2021) period compared to the control (Figs. 3b, 3c). ^{137}Cs uptake by leaves and green shoots of lingonberry generally followed the same pattern, with higher levels of ^{137}Cs TF_{ag} on treatment plots in the first two years of the study and about 40 % lower levels in the last three years, with no effect observed during the years 2014–2016 (Fig. 2e). The ^{137}Cs uptake by bilberry and lingonberry plants sampled prior to fertilisation on the EDTA treatment plot in April was approximately three times higher than in the control (Figs. 2b, 2e). Consequently, ^{137}Cs uptake by bilberry and lingonberry plants on the EDTA treatment plots was consistently higher to that in the control, and the effect of this treatment was delayed until 2018 (Figs. 2b, 2e, 3b, 3e). The decline of ^{137}Cs TF_{ag} values for bilberry and lingonberry plants over the study period on EDTA plots was two to three times faster than in the control plots (Table 2).

3.5.2. Rowan, alder buckthorn, birch and oak

The EDTA treatment generally had no effect on ^{137}Cs TF_{ag} in leaves and green shoots of the rowan and oak trees. However, during 2014–2016, it did increase ^{137}Cs uptake by about 50 % in rowan. In contrast, EDTA fertilisation had a negligible effect on ^{137}Cs TF_{ag} from soil to the leaves and shoots of alder buckthorn and birch, which decreased only marginally (38–44 %) although by the end of the study period, although non-significantly (Figs. 2a, 2d, 2f, 2g, 3a, 3d, 3f, 3g). Characteristically, the decline ratios of ^{137}Cs uptake by tree species under EDTA treatment and in the control plots, except for oak, were generally similar and close to one, indicating no changes over the years of study and no effect of soil fertilisation with micronutrients (Table 2).

4. Discussion

We found that applying three different fertilizers had a direct effect on the uptake of ^{137}Cs by young leaves and green shoots of the most common dwarf shrubs and tree species, confirming our initial hypothesis. However, the effectiveness of radionuclide transfer inhibition from soil to plants depends on the type of fertilisers.

Fertilisation with KCl was an effective countermeasure and reduced ^{137}Cs in both shrub and tree species. The effect of KCl treatment became more evident in the later years of the study, while the effect of K application in the beginning of the experiment was hidden by high ^{137}Cs uptake in KCl treatment plots in April 2012, prior experiment initiation, at least for bilberry and lingonberry. However, the more pronounced decrease in ^{137}Cs uptake on KCl treatments compared to the control plot indicates its relative high efficiency in reducing ^{137}Cs uptake.

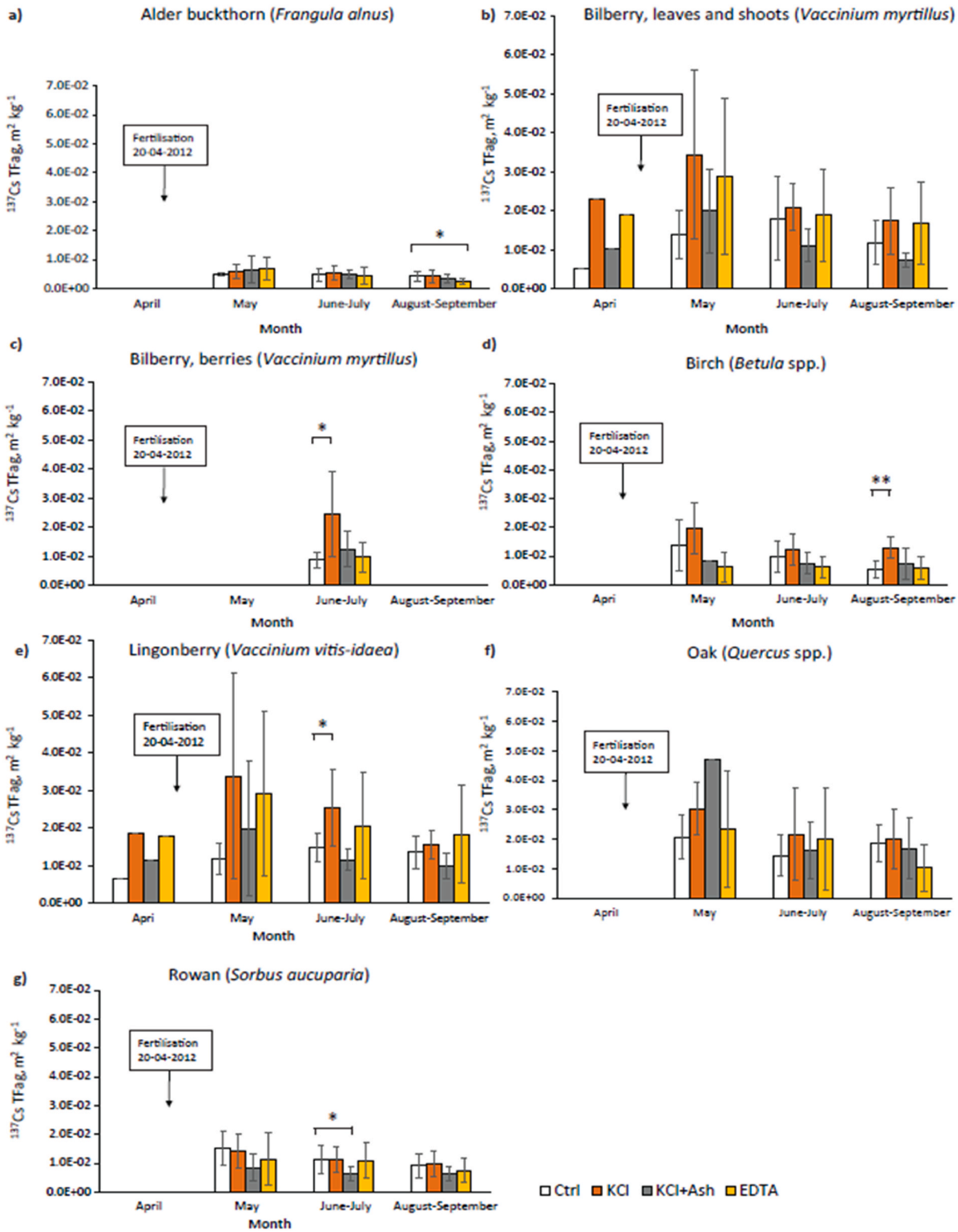

Fig. 2. Aggregated transfer factor (TF_{ag}, m^2 kg^{-1}) for ^{137}Cs in young shoots and leaves of dwarf shrubs and trees in the first growing season after treatment (April–September 2012) (n = 4). (**a**) alder buckthorn (*Frangula alnus* Mill.), (**b**) bilberry (*Vaccinium myrtillus* L.) leaves and shoots, (**c**) bilberry berries, (**d**) birch (*Betula* spp.), (**e**) lingonberry (*Vaccinium vitis-idaea* L.), (**f**) oak (*Quercus* spp.) and (**g**) rowan (*Sorbus aucuparia* L.). Significance levels are marked as follows: $p \leq 0.01$ (**), $p \leq 0.05$ (*). In April 2012, samples were taken prior to fertilisation.

The effect of K might be limited because of the high initial K content in the soils. It is generally accepted that a significant decrease in ^{137}Cs uptake by plants can be expected as a result of K fertilisation of mineral soils with exchangeable K content ≤20 mg kg^{-1} soil (Bilo et al., 1993). Soils in our experiment had a relatively high initial K content (70.3 mg kg^{-1}), corresponding to approximately 90 kg K per ha^{-1} at 15 cm depth, assuming a soil bulk density of 0.86 g/cm^3.

While a one-time application of 100 kg KCl per ha^{-1} may have been

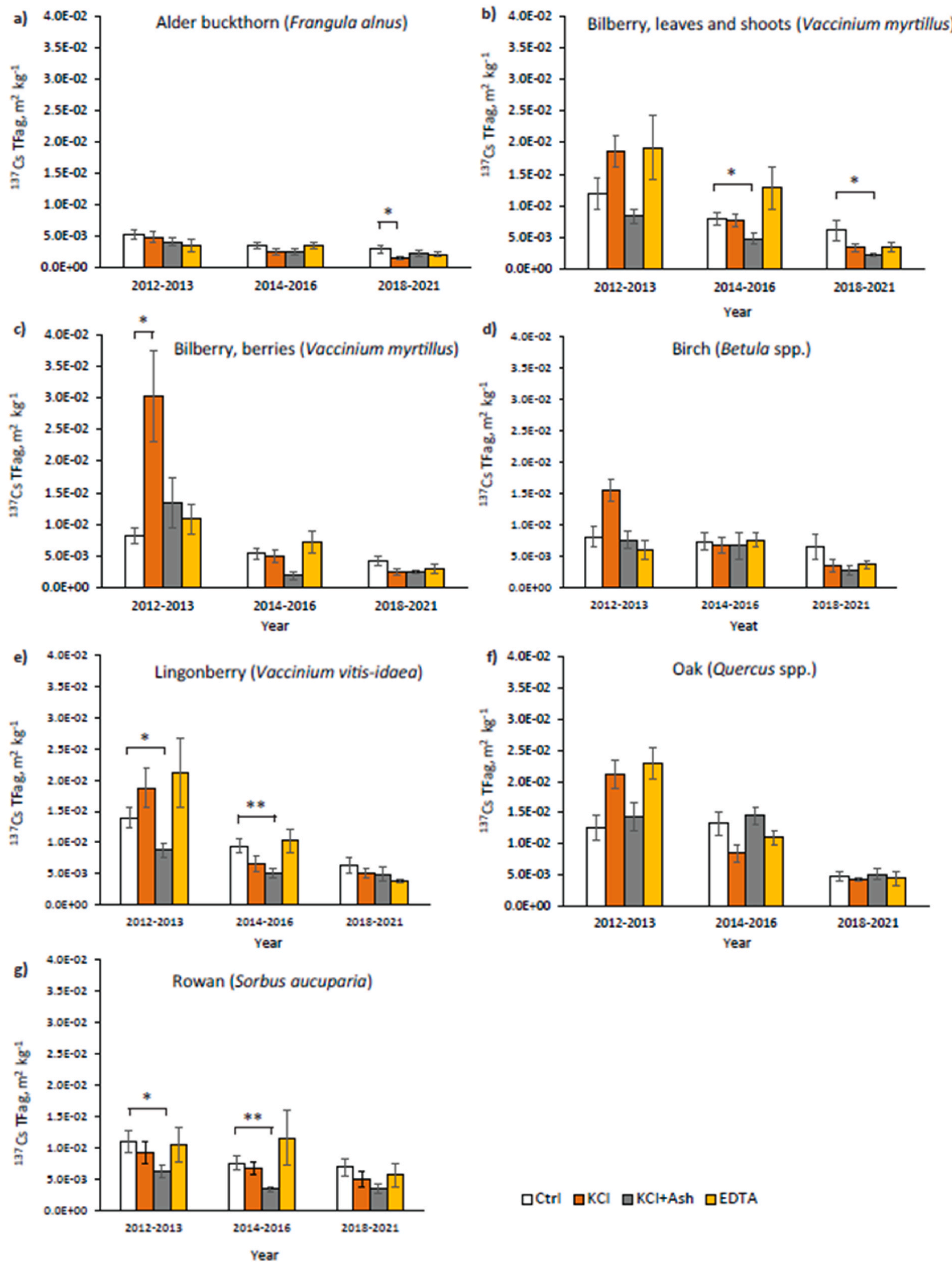

Fig. 3. Aggregated transfer factor (TF_{ag}, $m^2\ kg^{-1}$) for ^{137}Cs in young shoots and leaves of dwarf shrubs and trees in the 10-year study period (2012–2021) (n = 4). (**a**) alder buckthorn (*Frangula alnus* Mill.), (**b**) bilberry (*Vaccinium myrtillus* L.) leaves and shoots, (**c**) bilberry berries, (**d**) birch (*Betula* spp.), (**e**) lingonberry (*Vaccinium vitis-idaea* L.), (**f**) oak (*Quercus* spp.) and (**g**) rowan (*Sorbus aucuparia* L.). Significance levels are marked as follows: $p \leq 0.01$ (**), $p \leq 0.05$ (*).

insufficient to reduce ^{137}Cs uptake in the heavily contaminated soil, studies in Swedish forest sites with lower ^{137}Cs deposition (Rosén et al., 2011) showed that the same application decreased ^{137}Cs uptake by half. However, the ratio of ^{137}Cs to available K in those soils was lower than in our study, suggesting that the lack of significant effect in our study may be due to the small change in concentration ratio between plant-available ^{137}Cs and the added K.

Soils in the study area are still highly contaminated with ^{137}Cs (~250 kBq m^{-2}), but a significant proportion (up to 80%) has likely been bound to organic and mineral components, making it less available for exchange (Guivarch et al., 1999). The mobility of ^{137}Cs decreases over time (Roig et al., 2007), with the time elapsed since the 1986 fallout likely being the primary factor influencing its migration and fixation in soil.

The higher mobility of ^{137}Cs in soil, which makes it more available for plant uptake, may also contribute to the insufficient effect of K application on reduction of ^{137}Cs uptake by forest plants.

In soils with low ion exchange capacity, like those in our study area, the addition of KCl can mobilize ^{137}Cs by displacing other ions from soil surface sites, making it more readily available for plant uptake (Folder and Christenson, 1959; Grauby et al., 1990). The soddy podzolic soils in our study area, near the CNPP, have low cation exchange capacity and low pH levels. When K fertiliser is applied, it can displace other ions from the soil, releasing Cs^{+} ions that are more readily taken up by plants. This likely occurred in our study, where KCl fertiliser application mobilized ^{137}Cs through ion exchange reactions, counteracting the effects of fertilisation, especially in the first few years after treatment.

Fertilisation of soil with KCl in combination with wood ash was the most effective treatment and substantially reduced ^{137}Cs in both shrub and tree species, except for oak. Similar to KCl, the effect of the KCl + Ash treatment became more evident during the intermediate (2014–2016) and late (2018–2021) periods. For bilberry and lingonberry, the effect of the KCl + Ash treatment at the beginning of the experiment was obscured by high ^{137}Cs uptake in this plot prior to the start of the experiment, which may be attributed to factors other than fertilization. The more pronounced decrease in ^{137}Cs uptake on KCl + Ash treatments compared to the control plot also indicates its efficiency in reducing ^{137}Cs uptake.

Despite the same total amount of K applied, KCl + Ash had a stronger effect on ^{137}Cs uptake in tree and shrub species, likely due to the form of K in wood ash and its high pH value. These findings are in agreement with previous studies (Vetikko et al., 2003, 2010; Rantavaara and Aro, 2009; Vinichuk et al., 2023), which show that ash fertilisation reduces the ^{137}Cs transfer from soil to vegetation. The application of wood ash to forests can restore the pool of macronutrients, such as phosphorus and other nutrients in the soil (Tuyishime et al., 2022), which increases nutrient uptake by forest plants (e.g. *Vaccinium vitis-idaea* L) and decreases the uptake of ^{137}Cs (Levula et al., 2000).

Apart from increasing the abundance of nutrients in the humus layer, the use of wood ash in forest ecosystems also raises soil pH (Jacobson et al., 2004; Błońska et al., 2023) and reduces ^{137}Cs uptake. The wood ash we used was alkaline (pH $\approx$ 12), which likely contributed to raising soil pH and increasing the bioavailability of elements. Soil pH has been shown to negatively correlate with ^{137}Cs activity concentrations in, for example, oak shoots (Kanasashi et al., 2020), which explains the effect of the KCl + Ash treatment in the experiment.

Soil fertilisation with EDTA generally had no effect on reduction of ^{137}Cs uptake by the studied species. A non-statistically significant decrease of up to 40 % of ^{137}Cs TF_{ag} was observed only at the end of the experiment (2018–2021) for young leaves and green shoots of shrubs (bilberry plants and lingonberry) and birch trees. However, the ratio of ^{137}Cs TF_{ag} decline over the study period was about the same on both the EDTA and control plots, indicating no effect of fertilisation.

Sandy soils in north-western Ukraine are often deficient in Zn and Mn (Polupan, 1988). The EDTA treatment surprisingly increased ^{137}Cs transfer factors in young shoots and berries of bilberry and lingonberry for the first few years after treatment, before decreasing to a stable level.

Zn ions in soil are quickly absorbed by charged surfaces, with only a small amount absorbed by crops (Dubey et al., 2021). This suggests that ^{137}Cs may be more mobile and available in the soil, even when competing with other divalent cations. Our study is the first to investigate the impact of micronutrient fertilization on ^{137}Cs uptake by forest vegetation in heavily contaminated areas.

5. Conclusion

Our research suggests that a single application of KCl fertiliser to forests contaminated with radionuclides at a rate of 100 kg K ha^{-1} reduces ^{137}Cs uptake by most forest plants. While KCl fertiliser demonstrated limited effectiveness in reducing ^{137}Cs uptake, combining KCl with wood ash to supply 100 kg K ha^{-1} (with half from KCl and half from wood ash), can significantly reduce ^{137}Cs uptake in most tree and shrub species. However, the response to different fertilisers varied between species, likely due to differences in tree physiology and their mechanisms for transporting ^{137}Cs to young tissues. Soil fertilisation with EDTA does not seem to be a practical countermeasure, as the reduction in ^{137}Cs uptake by forest vegetation was only marginal and non-significant by the end of the study period.

The study's findings may be limited due to a small number of replicate plots and high variation in ^{137}Cs deposition values. Despite these limitations, the study provides a unique assessment of the long-term effects of different fertilisers on ^{137}Cs transfer in forest ecosystems. Further research is needed to understand the mechanism behind the observed fertiliser effects, particularly the potential role of K ions in releasing ^{137}Cs ions from soil binding sites, making them more available for plant uptake.

Declaration of generative AI and AI-assisted technologies in the writing process

Statement: During the preparation of this work, the authors did not use any Generative AI and AI-assisted technologies in the writing process.

CRediT authorship contribution statement

Mykhailo Vinichuk: Writing original draft, Writing – review and editing, Data curation, Visualization, Conceptualization, Funding acquisition, Project administration. **Yurii Mandro:** Investigation, Formal analyses, Data curation. **Julia Kyaschenko:** Writing – review and editing, Statistical analyses, Visualization, Validation. **Klas Rosén:** Supervision, Funding acquisition, Writing – review and editing.

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.

Acknowledgments

The project was funded by the Swedish Radiation Safety Authorities (grant no. 0113U004157 to MV, 2012), Royal Swedish Academy of Sciences (scholarship grant to MV), the Swedish University of Agricultural Sciences and Zhytomyr Polytechnic State University of Ukraine. We would like to thank Anatoliy Vyhovsky from Bazar Forestry of Zhytomyr region (Ukraine) for assistance and support in the field experiment.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.

org/10.1016/j.chemosphere.2024.143525.

Data availability

Data will be made available on request.

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