Journal of Environmental Management 336 (2023) 117609

Contents lists available at ScienceDirect

Journal of Environmental Management

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

Research article

Soil fertilisation with ^{137}Cs-contaminated and uncontaminated wood ash as a countermeasure to reduce ^{137}Cs uptake by forest plants

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ARTICLE INFO

Keywords:
Radiocaesium
Potassium
Wood ash
Forest vegetation
Ukrainian Polyssia
^{137}Cs uptake

ABSTRACT

The purpose of present study was to find out whether wood ash with a high pH value and neutralizing capacity reduces ^{137}Cs uptake by forest plants many years after the radionuclide fallout. The effects of one-time point fertilisation with ^{137}Cs-contaminated and uncontaminated wood ash alone or in combination with KCl on ^{137}Cs transfer from soil to young leaves and green shoots of various dwarf shrubs and tree species were examined in a long-term fertilisation experiment (2012–2021) conducted in Bazar mixed forest, around 70 km from Chernobyl nuclear power plant. The results indicated minor effects of soil fertilisation, although there were differences between ^{137}Cs uptake by species and years. Soil amendment with ^{137}Cs-contaminated wood ash generally did not affect ^{137}Cs uptake by young shoots and leaves of plants over the growing season in the first year and only slightly decreased T_{ag} for ^{137}Cs in the following years. The effect of a single application of ^{137}Cs-uncontaminated wood ash on reducing ^{137}Cs uptake by plants was generally negligible. Application of ^{137}Cs-contaminated wood ash in combination with KCl reduced plant ^{137}Cs uptake by about 45%, however, such reduction was only significant in some years for bilberry berries, young leaves and green shoots of lingonberry and alder buckthorn. Thus application of wood ash to ^{137}Cs-contaminated forest soil many years after radionuclide fallout generally does not reduce ^{137}Cs uptake by forest vegetation in a mixed forest ecosystem and this countermeasure should be applied with caution.

1. Introduction

Increased use of biofuels, including from organic materials and wood residues from forests, is leading to increased production of wood ash (Huotari et al., 2015). Wood ash is commonly used as a fertiliser in boreal forests, as it efficiently counteracts acidification and nutrient losses (Pitman, 2006). There may also be other benefits of adding wood ash to forests, e.g. a reduction in radiocaesium (^{137}Cs) concentration in plants (Levula et al., 2000).

Today, some 36 years after the release of radionuclides from the Chernobyl nuclear power plant, large areas of surrounding land are still heavily contaminated with ^{137}Cs, which is considered to be the most common dose-forming fission product. Most of the accumulated fallout of ^{37}Cs and radiostrontium (^{90}Sr) is still present in forests of Ukrainian Polyssia, where the level of ^{137}Cs in soil and vegetation is still very high. Understorey vegetation, edible mushrooms and berries contain high levels of ^{137}Cs, and therefore large animals such as moose and wild boar have elevated values (Ŝprem et al., 2013; Gulakov, 2014). People living in the area and consuming forest products are also exposed to high ^{137}Cs levels (Sartayev et al., 2021). Hence, there is a need to identify measures that reduce uptake of ^{137}Cs by forest plants.

Bioash and especially wood ash often have a relatively high content of potassium (K) (3–6% of dry matter) compared with other ash types (Nilsson, 2001; Pitman, 2006). Since K and Cs ions have similar chemical properties, an addition of K ions with wood ash may result in plants taking up K, and not Cs ions. Therefore ash application to forest soil could be used as a measure to reduce uptake of ^{137}Cs in woodland plants consumed by humans and wild animals. Previous studies assessing the effect of addition of wood ash on the content of ^{137}Cs in forest plants have focused on specific plant species (Levula et al., 2000), on plants in drained peatlands (Vetikko et al., 2010) or on field vegetation, needles and twigs of trees in coniferous forests (Högbom and Nohrstedt, 2001). The results obtained in those studies suggest that the content of ^{137}Cs in berries, mushrooms and tree parts is generally unaffected by application

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https://doi.org/10.1016/j.jenvman.2023.117609
Received 8 December 2022; Received in revised form 17 February 2023; Accepted 25 February 2023
Available online 4 March 2023
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of contaminated ash. Other investigations have shown that K fertilisation (usually as KCl) can reduce uptake of ^{137}Cs in forest plants (Rosén and Vinichuk, 2014) and thus potentially also the levels of ^{137}Cs in animals feeding on these plants (Levula et al., 2000; Kaunisto et al., 2002).

The present study is intended to highlight the knowledge gaps that exist regarding the time of fertilizers application since the radionuclide fallout (1) and regarding soils dominating in forest ecosystems in Ukraine (2). The aim was to investigate: 1) the effect of adding ^{137}Cs-contaminated or uncontaminated wood ash to ^{137}Cs-contaminated forest soil on uptake of ^{137}Cs in forest floor plants; and 2) whether a combination of ^{137}Cs-contaminated wood ash and conventional K fertiliser (as KCl) is more effective in reducing ^{137}Cs uptake by forest plants than wood ash alone.

The specific objective was to investigate the effectiveness of a single application of ^{137}Cs-contaminated wood ash (along or without K fertiliser KCl) and a single application of uncontaminated wood ash in reducing the content of ^{137}Cs in young leaves and green shoots of small woody, evergreen dwarf shrubs (bilberry (*Vaccinium myrtillus* L.) and lingonberry (*Vaccinium vitis-idaea* L.)) and trees (birch (*Betula* spp.), rowan (*Sorbus aucuparia* L.), oak (*Quercus* spp.) and alder buckthorn (*Frangula alnus* Mill.)). Young leaves and green shoots are the most metabolically active plant tissues and have relatively high K requirements, and therefore ^{137}Cs, which follows the metabolic pathway of K, can be expected to accumulate in these plant organs in relatively large amounts.

The results obtained were used to assess addition of ^{137}Cs-contaminated or uncontaminated ash as a measure for reducing plant uptake of ^{137}Cs in areas affected by radioactive fallout.

The hypotheses tested were that:

(a) Adding K fertiliser (KCl) in combination with ^{137}Cs contaminated or uncontaminated wood ash reduces plant uptake of radioactive ^{137}Cs compared to adding wood ash alone, due to the combined effect of K in the form of KCl and the liming effect of the ash.
(b) Applying wood ash with a high or low content of ^{137}Cs makes little difference to plant uptake of ^{137}Cs, because of supply of available K in the ash.

2. Materials and methods

2.1. Site description

A field experiment was established in Bazar forest within the Drevlianskyi Natural Reserve (51°05′35″N, 29°18′56″E), Zhytomyr region, Ukraine, about 70 km from the Chernobyl nuclear power plant. In 2012–2014, ^{137}Cs background radioactivity in the area ranged between 177 and 355 kBq m^2 with a mean value of 273 kBq m^2. The major soil type is a typical forest soil, a soddy podzolic soil developed on glacial-fluvial sediments and characterised by low cation exchange capacity (1.1–11.0 meq 100 g^{-1} of soil) which makes ^{137}Cs easily plant available.

The thickness of the organic horizon generally does not exceed 3–5 cm. The main chemical characteristics of the 0–15 cm soil layer before the start of the experiment were as follows (mean ± SD, n = 14): pH 4.25 ± 0.47, total carbon (C) 2.96 ± 1.93%, total nitrogen (N) 0.12 ± 0.09%, C/N ratio 24.76 ± 2.78, ammonium lactate extractable K (K-AL) 7.03 ± 6.36 mg 100 g^{-1}, calcium (Ca-)AL 16.82 ± 19.22 mg 100 g^{-1}, hydrochloric acid-extractable K (K–HCl) 15.63 ± 6.31 mg 100 g^{-1}, Ca–HCl 22.85 ± 21.70 mg 100 g^{-1}.

The dominating tree species at the study site is Scots pine (*Pinus sylvestris* L.), with some intermixed birch. Other common tree species are rowan, oak and alder buckthorn. The most common dwarf shrubs are bilberry, lingonberry, bracken (*Pteridium aquilinum* (L.) Kuhn) and heather (*Calluna vulgaris* L.). The forest floor and tree bark are partly covered with various mosses and lichens, mainly *Dicranum polysetum* Sw. and *D. scoparium* Hedw. The stand is approximately 60–80 years old.

2.2. Experiment layout

In experiments carried out 2012–2021, four treatments were compared: 1) Control (non-fertilised); 2) ^{137}Cs-contaminated wood ash (^{137}CsAsh); 3) uncontaminated wood ash (Ash); and 4) KCl in combination with ^{137}Cs-contaminated wood ash (supplying 50 kg K ha^{-1} from KCl and 50 kg K ha^{-1} from wood ash) (KCl+^{137}CsAsh). Samples of plant species were taken from each experimental plot.

The ^{137}Cs-contaminated loose ash used in the experiment originated from combusted wood residues from thinning, clearing and other cuttings in a neighbouring area in the same forest. The uncontaminated wood ash was a mixture of loose ash originating from wood-burning stoves (grate ash) and combusted wood residues from a ^{137}Cs-uncontaminated neighbouring area. The ^{137}Cs-contaminated and uncontaminated ash both had pH 12.8, total phosphorus (P) content 0.9%, total K content 3.1% and had an activity concentration (AC) of ^{137}Cs of 17 200 and 46.0 Bq kg^{-1}, respectively. The only total concentration of potassium was measured in ashes, assuming that both potassium minerals in wood ashes and potassium fertilisers are highly soluble in water (Etitgni and Campbell, 1991; Erich, 1991).

Potassium chloride with a content of 50% K was used as the K fertiliser. The products were spread by hand on the forest floor once in April 2012, at a rate corresponding to approximately 100 kg ha^{-1} K. Each treatment had four replicates, resulting in a total of 16 plots (4 treatments × 4 replicates). The treatments were randomly allocated to 200 m^2 experimental plots (20 m × 10 m) within an area of about 0.32 ha.

2.3. Sampling and treatment

Two soil samples were taken from each of 16 plots in 2012 and 2014, using a cylindrical steel core with diameter 57 mm and working section length 150 mm. For spectrometric measurements and chemical analyses, soil was air-dried to constant weight, milled, sieved through a 2 mm mesh to produce a homogeneous material, and weighed again. For chemical analysis, 10 g of air-dried soil was used.

In 2012, aboveground green parts of bilberry, lingonberry and young shoots of trees were taken every month during May–September. In the following years (2013, 2014, 2015, 2016, 2018 and 2021), samples were taken once per vegetation season (late June or early July). Collected plant samples (about 2000 in total) were air-dried, cut into smaller pieces, mixed thoroughly, weighed and placed in standard 35-L plastic tubes for gamma spectrometric measurements. Bilberry berries were collected occasionally when available, dried at 65 °C, weighed and analysed for ^{137}Cs AC.

2.4. Radiometry and data treatment

Activity concentration of ^{137}Cs (Bq kg^{-1} dry weight, d. w.) was measured using HPGe (High-purity germanium) and NaI detectors to achieve an error below 5%, but not for longer than 24 h. All results were decay-corrected to the date of sampling. Data were processed using WinDAS™ (Windows Data Acquisition System) and Apex-Gamma™ software. Basic statistics and a two-sample *t*-test were performed using Minitab® 19.2020.1 software.

To test the effect of different fertilisers on each plant species analysed over the study period, a repeated measures mixed effect model was used, with sampling plot as a random factor (n = 56) and with plot treatment (fertilised or control), log-transformed ^{137}Cs AC in plant samples, year and interaction between plot treatment and year as candidate explanatory variables. The model was fitted using the R-package nlme (Pinheiro and Bates, 2022).

Soil contamination density ($A_{s,\ Bq\ m^{-2}}$) was calculated as:

$$A_s = \frac{(A_m \times m)}{S} \quad (1)$$

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

Aggregated transfer factor (T_{ag}, m^2 kg^{-1}) for ^{137}Cs transfer from soil to plant material was calculated as:

$$T_{ag} = \frac{A_m}{A_s} \quad (2)$$

where A_m is activity concentration of ^{137}Cs per unit dry weight of plant material on sampling date (Bq kg^{-1}); A_s is the average for years 2012 and 2014 level of soil contamination with ^{137}Cs (Bq m^{-2}).

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

3. Results

The average ^{137}Cs deposition in the experimental plots was 272 kBq m^{-2} (range 141–479 kBq m^{-2}), with only limited variation within three of the treatments (Control, ^{137}CsAsh, Ash) (Appendix A). Treatment KCl+^{137}CsAsh had a higher ^{137}Cs level (342 kBq m^{-2}). Generally, the level of ^{137}Cs within the experimental area is still relatively high, with nearby territories classified as zone 2 (unconditional (mandatory) resettlement zone) in 1986 owing to ^{137}Cs levels exceeding 555 kBq m^{-2} (Verkhovna Rada, 1991).

3.1. ^{137}Cs activity concentration

Estimated ^{137}Cs AC in bilberry plants from control plots varied between 1.01 and 3.17 kBq kg^{-1} for young leaves and green shoots, and between 0.82 and 2.21 kBq kg^{-1} for berries. The values were highest at the beginning of the experiment and decreased gradually over the study period, by a factor of 2.3 for plants and 2.5 for bilberry berries (Appendix B). In lingonberry, ^{137}Cs AC in leaves and green shoots also decreased over time, by factor of around 2.8, and ranged between 1.18 and 3.27 kBq kg^{-1} during the study period. ^{137}Cs AC in leaves and green shoots of rowan and birch trees decreased from 2.80 to 1.95 kBq kg^{-1}, respectively, in 2012 to about 1.50 kBq kg^{-1} in 2021, i.e. the decrease was less pronounced for birch trees. Oak leaves and green shoots had the highest ^{137}Cs AC at the beginning of the experiment (4.09 kBq kg^{-1}), while alder buckthorn leaves and green shoots had the lowest (1.18 kBq kg^{-1}) (Appendix B). ^{137}Cs AC in oak and alder buckthorn samples decreased by a factor of 2 and 4.2, respectively, during the course of the experiment.

Fertilisation with ^{137}Cs-contaminated wood ash, with or without KCl, or with uncontaminated wood ash generally did not affect ^{137}Cs AC in leaves and green shoots of plants in the beginning of the experiment (2012), but resulted in a noticeable but not always significant reduction at the end of the experiment (2021) (Appendix B). ^{137}CsAsh was the most effective treatment, resulting in the highest (up to 100-fold) reduction in ^{137}Cs AC in bilberry, rowan and birch over the study period. The other treatments resulted in a slightly smaller decline in ^{137}Cs activity (5- to 7-fold) in fertilised plants over time compared with the control.

3.2. Aggregated transfer factor

Aggregated transfer factors for ^{137}Cs in young shoots and leaves of dwarf shrubs growing in control plots were within the range 12.5–13.3 $\times 10^{-3}$ m^2 kg^{-1} in 2012 and decreased to about 5.0–5.9 $\times 10^{-3}$ m^2 kg^{-1} in 2021 (Appendix C). The T_{ag} for ^{137}Cs in trees growing in control plots was within the range 5.4–15.9 $\times 10^{-3}$ m^2 kg^{-1} in 2012 and decreased to about 2.37.3 $\times 10^{-3}$ m^2 kg^{-1} in 2021. On average, ^{137}Cs transfer from soil to young leaves and green shoots of plants growing in control plots during the whole study period declined gradually, by a factor of 2 (Table 1). Estimated T_{ag} for ^{137}Cs in plants growing in fertilised plots declined more rapidly over time. For some plant species (bilberry, rowan, birch), ^{137}Cs uptake over the nine years of the experiment decreased by around 9-fold, while other species (lingonberry, alder buckthorn, oak) showed a 4- to 7-fold reduction (treatment with ^{137}Cs-contaminated wood ash) (Table 1). For plants growing in plots fertilised with uncontaminated wood ash and ^{137}Cs-contaminated wood ash in combination with KCl, a 3- to 4-fold decline in ^{137}Cs uptake during the study period was observed (Table 1).

There was a significant effect of fertilisation with ^{137}CsAsh, Ash and KCl+^{137}CsAsh in reducing ^{137}Cs uptake by bilberry plants ($p = 0.01$, 0.03 and 0.04, respectively), but the effect was not significant for other species (Table 1).

3.3. Fertilisation with ^{137}Cs-contaminated wood ash (^{137}CsAsh)

3.3.1. Bilberry and lingonberry

In general, soil amendment with ^{137}Cs-contaminated wood ash did not affect ^{137}Cs uptake by young shoots and leaves of bilberry (plants and berries) or lingonberry over the growing season (April–September) in the first year (2012) (Fig. 1) but it decreased T_{ag} for ^{137}Cs by 50–60% in the following years (2015–2021) (Fig. 2).

The strongest effect of ^{137}CsAsh fertilisation (reduction up to 70%) was seen in year 9 after fertilisation (2021), but the overall effect of fertilisation with ^{137}Cs-contaminated wood ash over the experimental period was not significant ($p \geq 0.5$).

3.3.2. Rowan, alder buckthorn, birch and oak

Young shoots and leaves of rowan, birch, alder buckthorn and oak growing in the ^{137}CsAsh plots showed slightly higher ^{137}Cs uptake compared with the control in the first year after fertilisation (Fig. 1) and substantially lower values (by 50–60%) in the following years (Fig. 2).

Table 1
Decrease in aggregated transfer factor (T_{ag}) for ^{137}Cs in forest plants over the study period (2012–2021).

Species	^{137}Cs T_{ag}, $\times 10^{-3}$ m^2 kg^{-1}		Decline, ratio[a]	P-value	^{137}Cs T_{ag}, $\times 10^{-3}$ m^2 kg^{-1}		Decline, ratio	P-value	^{137}Cs T_{ag}, $\times 10^{-3}$ m^2 kg^{-1}		Decline, ratio	P-value	^{137}Cs T_{ag}, m^2 kg^{-1}		Decline, times	P-value
	2012	2021			2012	2021			2012	2021			2012	2021		
	Control				^{137}CsAsh				Ash				KCl+^{137}CsAsh			
Bilberry	12.5	5.89	2.1	0.21	12.9	1.46	8.8	**0.01**	8.22	3.32	2.5	**0.03**	11.5	1.96	5.9	**0.04**
Lingonberry	13.3	5.02	2.7	**0.03**	15.3	2.76	5.5	0.11	14.1	4.08	3.4	0.08	11.2	3.23	3.5	0.06
Rowan	11.1	6.05	1.9	0.19	16.4	1.59	9.2	0.07	11.3	3.16	3.6	**0.001**	6.89	3.21	2.1	0.30
Birch	7.24	7.30	1.0	0.98	11.4	1.19	9.5	**0.02**	8.78	6.19	1.4	0.41	11.4	3.37	3.4	**0.02**
Alder buckthorn	5.38	2.28	2.3	**0.03**	6.69	0.90	6.7	0.06	4.03	1.57	2.6	**0.03**	3.41	1.24	2.7	0.09
Oak	15.9	3.76	4.2	**0.04**	12.7	2.97	4.3	0.12	20.9	4.50	4.5	–	10.7	2.13	5.0	0.18
Bilberries	10.0	4.12	2.5	0.09	11.82	1.60	7.4	**0.04**	9.13	2.50	3.7	0.20	11.5	2.20	5.2	**0.04**

[a] Calculated as mean^{137}Cs TF_{ag}, m^2 kg^{-1} in 2012 divided by mean^{137}Cs TF_{ag}, m^2 kg^{-1} in 2021. The obtained values are probably overestimated as^{137}Cs TF_{ag} were calculated on the average (years 2012–2014) deposition level of^{137}Cs in soil.

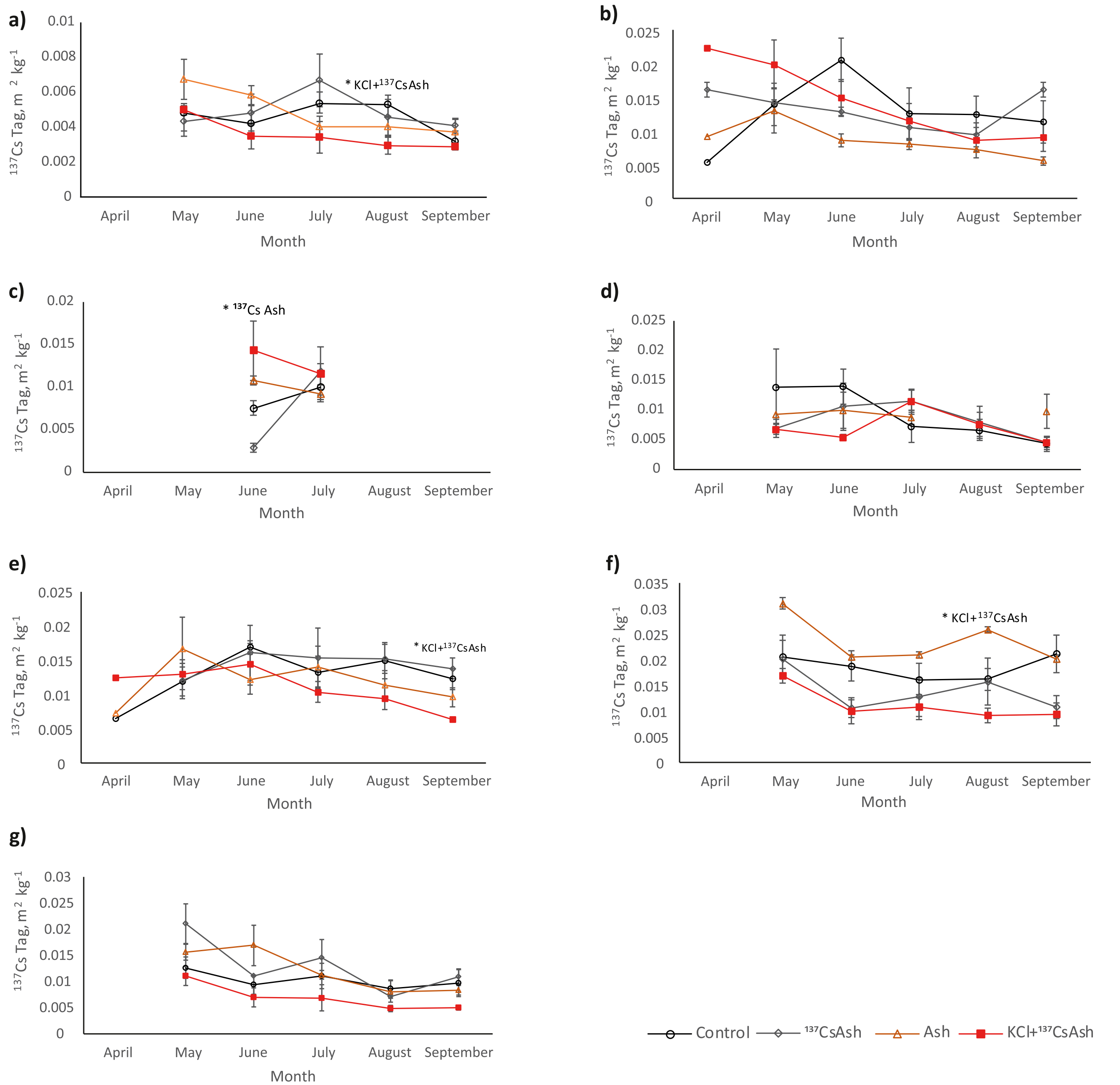

Fig. 1. Aggregated transfer factor (T_{ag}, $m^2\ kg^{-1}$) for ^{137}Cs in young shoots and leaves of dwarf shrubs and trees in the growing season April–September 2012 (n = 4). * indicates significant difference between treatment and control ($p < 0.05$). (a) alder buckthorn (*Frangula alnus* Mill.), (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.).

Apart from oak leaves and shoots, the effect of application of ^{137}Cs-contaminated wood ash to soil was strongest during the last years of the study ($p < 0.5$).

3.4. Fertilisation with uncontaminated wood ash (Ash)

3.4.1. Bilberry and lingonberry

Fertilisation with uncontaminated wood ash had no effect on ^{137}Cs uptake by bilberry and lingonberry plants in April and May in the first year of the experiment (Fig. 1). Between June and September of the first year, ^{137}Cs uptake by bilberry plants growing in Ash plots was 40–50% lower than in plants in unfertilised (control) plots, while lingonberry plants did not show any difference in ^{137}Cs accumulation (Fig. 1). In the following years, bilberry and lingonberry plants in Ash plots generally showed 30–40% lower ^{137}Cs transfer than those in control plants, but the difference was not significant ($p \geq 0.5$).

3.4.2. Rowan, alder buckthorn, birch and oak

During the first year, ^{137}Cs uptake by leaves and shoots of tree species growing in plots fertilised with uncontaminated ash was similar or higher than that in control plots (Fig. 1). In the following years (2013–2021), ^{137}Cs uptake by leaves and shoots of oak in Ash plots was similar to that in control plots, while ^{137}Cs transfer to leaves and shoots of rowan, birch and alder buckthorn was reduced by 20–30% in fertilised plots, although the difference was not significant ($p \geq 0.5$) (Fig. 2).

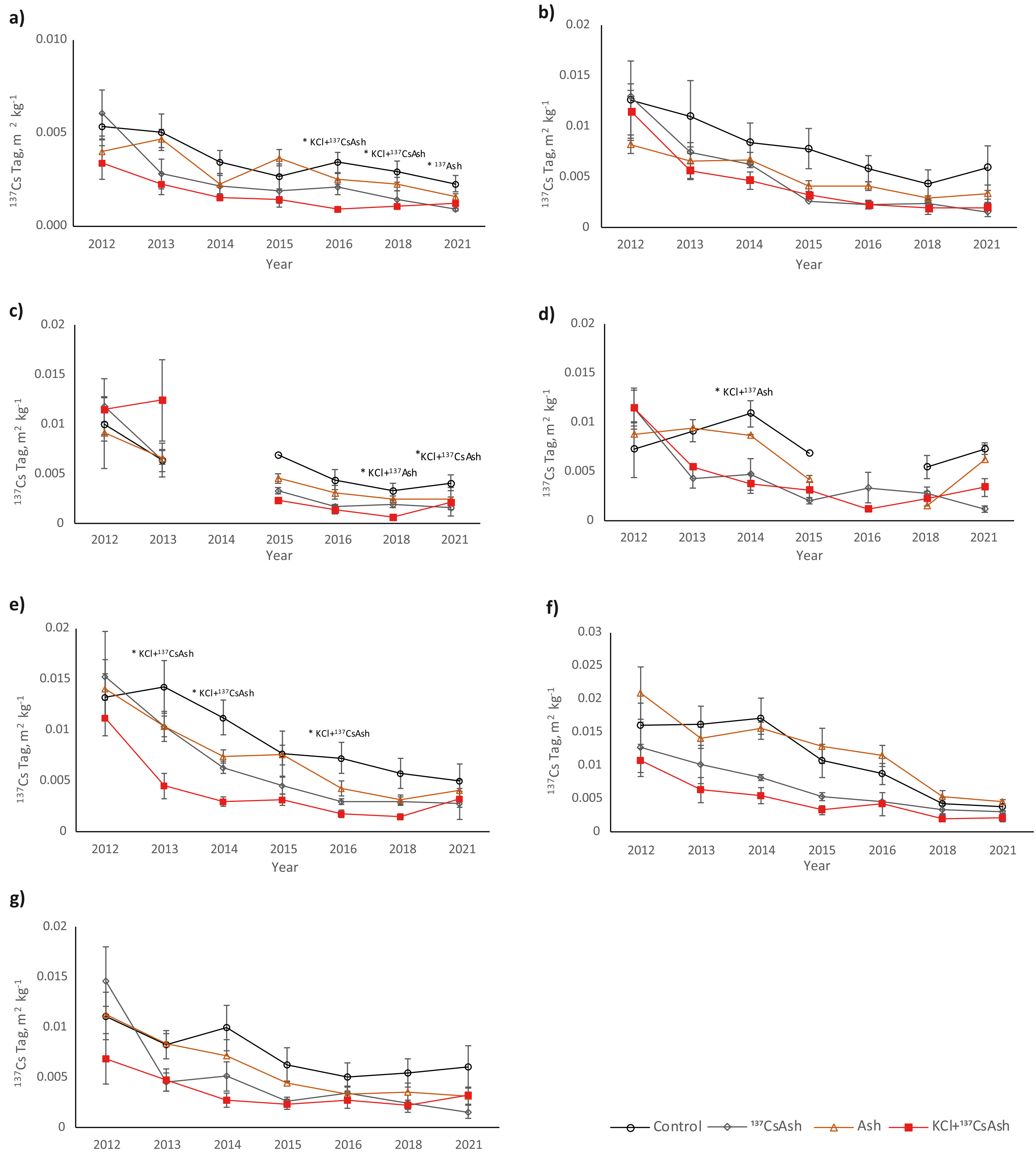

Fig. 2. Aggregated transfer factor (T_{ag}, $m^2\ kg^{-1}$) for ^{137}Cs for young shoots and leaves of dwarf shrubs and trees over the study period (2012–2021) (n = 4). * indicates significant difference between treatment and control ($p < 0.05$). (a) alder buckthorn (*Frangula alnus* Mill.), (b) bilberry (*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.).

3.5. Fertilisation with KCl in combination with ^{137}Cs-contaminated wood ash (KCl+^{137}CsAsh)

3.5.1. Bilberry and lingonberry

Application of K fertiliser in combination with ^{137}Cs-contaminated wood ash resulted in higher ^{137}Cs uptake by bilberry and lingonberry plants at the beginning of the growing season (April–May) in the first year, followed by lower (30–50%) uptake already at the end of the season (August–September) (Fig. 1). For lingonberry plants, the reduction in ^{137}Cs uptake was found to be significant (up to 60%) already in September 2012 (Fig. 1). In KCl+^{137}CsAsh-fertilised plots, uptake of ^{137}Cs by leaves and green shoots of bilberry and lingonberry over the study period was reduced by a factor of about 2, but the effect was only significant ($p < 0.05$) for lingonberry plants in 2013, 2014 and 2016 (Fig. 2). Bilberry berries from KCl+^{137}CsAsh plots showed significantly lower ^{137}Cs uptake in the 2016 and 2018 (Fig. 2).

3.5.2. Rowan, alder buckthorn, birch and oak

There was no effect of KCl+^{137}CsAsh fertilisation on birch tree ^{137}Cs uptake during the first year of the experiment, while ^{137}Cs transfer to rowan, alder buckhorn and oak was reduced by 30–40% already in the second half of the growing season (July–September 2012) (Fig. 1). For alder buckhorn and oak leaves and shoots, the ^{137}Cs reduction was significant ($p = 0.023$ and 0.038, respectively) (Fig. 1). In the following years, trees growing in KCl+^{137}CsAsh-fertilised plots had generally 50–60% lower T_{ag} for ^{137}Cs compared with trees growing in control plots (Fig. 2).

4. Discussion

There was only minor variation in soil concentration of ^{137}Cs within control, ^{137}CsAsh and Ash treatments plots (~250 kBq m^{-2}), whereas two of four plots in treatment KCl+^{137}CsAsh had higher soil ^{137}Cs concentration (469 kBq m^{-2}) (see Appendix A). This difference is probably related to unevenness of spatial distribution of radioactive fallout within the forest floor. Due to this variation in ^{137}Cs deposition within the sampling areas, the measurement results were compared using the values of T_{ag} for ^{137}Cs.

Although ^{137}Cs AC in the ash used in experiments was up to 10-fold higher than that in the original wood, application of ^{137}Cs-contaminated wood ash made only a negligible contribution to the soil ^{137}Cs inventory, since 3000 kg ha^{-1} of contaminated wood ash with ^{137}Cs AC of 17.2 kBq kg^{-1} contributed about 5.16 kB m^{-2} of radioactivity to the soil.

^{137}Cs Tag values varied between plant species studied being generally highest for leaves and shoots of blueberry, lingonberry and oak and lowest for alder buckthorn and birch. However, the difference in ^{137}Cs Tag values between species was not found to be significant due to large variation within replicates and between years.

The ^{137}Cs-contaminated and uncontaminated wood ash products were both applied at a rate of 3000 kg ha^{-1}, which corresponded to 100 kg K ha^{-1}. This amount of ash is unlikely to affect biodiversity or lead to a decrease in species abundance in the forest (Arvidsson, 2001).

Interpretation of T_{ag} values was rather complicated, since the effect of wood ash application was somewhat indistinct. The ^{137}Cs-contaminated and uncontaminated wood ash used in this study were both alkaline (pH about 12) and both contained base cations, such as Ca. This gives wood ash liming properties that can counteract soil acidity by reducing soil pH and increasing the bioavailability of elements (Demeyer et al., 2001). Wood ash also increases the base cation content in the soil solution (from 30 to 90% according to Park et al. (2004), especially in the upper humus layers of the soil profile (Bang-Andreasen et al., 2021) and to a lesser degree in the mineral soil (Jacobson et al., 2004). This increase in base cations presumably has a liming effect on the soil, increasing pH and reducing total acidity in the upper organic soil layers. Thus, the decrease in ^{137}Cs uptake observed for some plant species in this study was probably related to the increased concentration of K and other soil nutrients (e.g. Ca, P) in the soil solution and to the ash-related increase in soil pH. The lack of pronounced effects may have been partly due to the liming effect of ash being short-lived (Scheepers, 2014).

The weak effects of ^{137}Cs-contaminated and uncontaminated wood ash in reducing ^{137}Cs uptake in forest plants are in good agreement with findings in a similar study on Swedish forests (Högbom and Nohrstedt, 2001), where no statistically significant effects of ^{137}Cs-contaminated wood ash application on ^{137}Cs activity in plants were found at six of seven sites. However, in the present study, the most pronounced decrease in ^{137}Cs uptake following wood ash application was seen in the treatment KCl+^{137}CsAsh. This was likely due to synergistic effects of the presence of K inhibiting ^{137}Cs uptake and to the liming effect of the ash. Addition of K is known to decrease ^{137}Cs uptake by trees (Komatsu et al., 2017; Kaunisto et al., 2002), forest dwarf shrubs and fungal fruiting bodies (Rosén et al., 2011).

An effect of combined K-wood ash fertilisation may be expected if the K concentration in the soil solution of K is below approximately 1 mmolL^{-1} (Smolders et al., 1997) or if the exchangeable K content is 20 mg kg^{-1} soil or less (Nisbet, 1995). Plant-available K content in soil within the experimental area is around 7.0 mg 100 g^{-1} soil, which can be considered insufficient, so an effect of K fertilisation could be expected. Based on field observations, we suggest that effect of soil amendment may be counteracted by the factor of time. Transfer of ^{137}Cs from soil to plants mainly depends on the clay content in soil, the exchangeable K content in the soil solution and the time after fallout (Absalom et al., 1999, 2001). Since our experimental soil was fertilised 36 years after fallout, ^{137}Cs had presumably become more strongly fixed and less available for uptake, even though the sandy soil at the site had a low content of clay and silt (6.9%). Some 10 years after the fallout, Chernobyl-derived ^{137}Cs in mineral soil was found to be significantly (by about a factor of two) more mobile than ^{137}Cs from the global fallout (Bunzl et al., 1995). In soils with low ion exchange capacity, a high K concentration following K addition may mobilise ^{137}Cs (Folder and Christenson, 1959), counteracting the effect of fertilisation. Soddy podzolic soils within the area around Chernobyl nuclear power plant are characterised by low cation exchange capacity (1.1–11.0 meq 100 g^{-1} of soil) and acid reaction of the soil solution.

A substantial (up to 70%) part of ^{137}Cs activity may be bound within organic matter (Koarashi et al., 2019), additionally, up to 50% of the total ^{137}Cs activity may be retained within fungal mycorrhizal structures (Vinichuk and Johanson, 2003) and forest vegetation (McGee et al., 2000). This fraction of ^{137}Cs activity becomes available for plant uptake due to recycling of ^{137}Cs in the organic surface horizons and transformation of litter within the F-layer (Mensah et al., 2021).

An uneven pattern of radioactivity within the forest floor and high variability in radioactivity data are additional sources of variation that complicate interpretation of the results and make the effect of forest soil fertilisation less consistent.

5. Conclusions

This study showed that may be difficult to inhibit ^{137}Cs transfer from soil to forest plants by adding ^{137}Cs-contaminated wood ash, alone or in combination with KCl, or non-contaminated wood ash many years after the initial fallout. The wood ash and K treatments tested here were found to have inconsistent and relatively small effects. One reason is that a relatively long time (36 years) has elapsed since the fallout and ^{137}Cs has presumably become more strongly fixed in soil and less available for uptake, even though soil concentrations of ^{137}Cs are still relatively high. Part of ^{137}Cs radioactivity in forest soil is likely to be fixed by soil fungi and vegetation, becoming available for uptake only after decomposition. A high K concentration in sandy soil after K fertilisation may also partly mobilise ^{137}Cs, counteracting the mitigating effect of fertilisation. Thus, addition of wood ash to forest soil as a practical countermeasure to reduce ^{137}Cs transfer from soil to forest vegetation in a long-term perspective should be used with caution. However, interpretation of the results is somewhat complicated due to well-known variation in ^{137}Cs deposition from the Chernobyl accident and consequently in ^{137}Cs radioactivity concentrations in forest plants. The lack of evidence of an effect of fertilisation may also be partly due to the low number of replicate plots used ($n = 4$). Thus further studies are required, since forest soil amendment with wood ash may be beneficial if appropriate rates of wood ash are applied.

Authorship contribution statement

Mykhailo Vinichuk: Conceptualisation, Methodology, Writing-

Reviewing and Editing; **Yurii Mandro**: Data curation, Investigation; **Julia Kyaschenko**: Visualisation, Software, Validation, Reviewing and Editing; **Klas Rosén**: Supervision, Reviewing 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.

Data availability

Data will be made available on request.

Acknowledgments

The project was funded by the Swedish Radiation Safety Authorities (grant # 0113U004157 to MV, 2012), Royal Swedish Academy of Sciences (scholarship grant to MV), Swedish University of Agricultural Sciences and Zhytomyr Polytechnic State University, Ukraine. We would like to thank Anatoliy Vyhovsky from Bazar Forest in 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.jenvman.2023.117609.

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