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Activity of ^{137}Cs and ^{40}K Isotopes in Pine (*Pinus sylvestris* L.) and Birch (*Betula pendula* Roth) Stands of Different Ages in a Selected Area of Eastern Poland

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Abstract: Research Highlights: a forest is an ecosystem that allows for the assessment of radioactive contamination of the environment over several decades. (1) Background and Objectives: measurements of the activity of the ^{137}Cs isotope in various elements of a forest ecosystem are one of the most important parameters in the assessment of radioisotope contamination. The translocation of ^{137}Cs in the environment is determined by the activity of the natural ^{40}K isotope in soil. The activities of ^{137}Cs and ^{40}K isotopes were assessed in two stands of *Betula pendula* and *Pinus sylvestris*, differing in age (30, 50, and 80 years old); (2) Materials and Methods: the research was conducted in one of the forest districts of eastern Poland. Wood, litter, and soil were collected for the tests from the sampling sites. The activity of ^{137}Cs and ^{40}K was determined using the γ -spectrometric method. Based on the activity of these isotopes in wood and soil, the values of translocation factors (TF) were determined; (3) Results: the highest activity of ^{137}Cs was recorded in the wood of the oldest stands, the growth period of which coincided with the time period of intensive testing with nuclear weapons. With the growing age of the stand, the value of TF ^{137}Cs increased, while the value of TF ^{40}K was not dependent on the age of the stands. Birch wood accumulated more ^{137}Cs and ^{40}K isotopes than pine wood. (4) Conclusions: the results show a much greater radioactive contamination of the environment in eastern Poland during the testing with nuclear weapons than after the Chernobyl nuclear reactor explosion. The greater accumulation of radiocesium in birch wood than in pine wood predisposes this species to be more useful when assessing the radioactive contamination of the environment from the past.

Keywords: radiocesium; potassium; wood; soil; litter; radioactive contamination; translocation factor

1. Introduction

The radioactive contamination of the environment that occurred in Europe during the second half of the 20th century was caused by nuclear weapons testing and the explosion of the Chernobyl nuclear reactor. Before 1999, more than 2500 trials with nuclear weapons had been performed worldwide, resulting in approximately 500 explosions in the atmosphere; most of them (about 90%) were executed between 1945–1962 [1]. The measurement of radioactive contamination of the environment with radioactive isotopes is the i.a. fallout of β -radioactive isotopes. Measurements of near ground-layer radioactivity conducted in Poland during the 1960s, exhibited particularly high values of radioactive isotope concentrations in total precipitation samples [2]. For example, in Poland, the average daily precipitation of β -radioactive isotopes during the years 1960–1963 varied within the range of 89–133 Bq/m², while in 1985, the value was 1 Bq/m². In 1986, following the nuclear disaster in Chernobyl, the concentration was 54 Bq/m², and during the following year, the precipitation returned to the levels observed prior to the accident [2].

^{137}Cs is one of the β -radioactive isotopes causing radioactive contamination of the environment. The half-life ($T_{1/2} = 30.1$ years) allows for the analysis of its activity in soil, even several decades after its release into the environment. Forest ecosystems play a crucial role in the adsorption and redistribution of the ^{137}Cs radionuclide. Compared to other ecosystems, in forests, it is possible to analyze the migration of radioactive contamination for several decades after the contamination [3,4]. Certain plant species that are characterized by a long life cycle and a wide range of occurrence, tolerant of various environmental conditions and showing the ability to accumulate pollutants in their tissues, are particularly useful in the assessment of environmental contamination [5]. The tree species widespread in forests and highly tolerant of climatic and habitat conditions are *Pinus sylvestris* L. [6] and *Betula pendula* Roth [7]. Both species accumulate the radioactive ^{137}Cs isotope [8].

After the Chernobyl disaster, numerous studies have investigated the accumulation of radioactive isotopes in the forest environment. They have highlighted the important role of forest litter and the surface layer of soil enriched with organic matter in the accumulation of the ^{137}Cs isotope [8–11]. Contaminated soil is the primary source of radionuclides for plants. Apart from soil contamination, there are also other factors determining cesium accumulation in trees and other plants, including species and age, agrochemical soil properties, topographic, meteorological, and ecological conditions [3,4]. An important factor influencing the level of ^{137}Cs accumulation in plants is the potassium content in the soil. Both cesium and potassium represent the elements of group I of the periodic table and are characterized by similar chemical properties; however, they behave competitively in relation to each other [12,13]. Unlike cesium, potassium is a natural component of the soil and has a radioactive ^{40}K isotope in its isotopic composition ($T_{1/2} = 1.32 \times 10^9$ years) [14]. In studies on the interrelationship between Cs and K in environmental samples, transfer coefficients are often taken into account [12,15,16]. They are considered one of the most important parameters in environmental safety assessment [15].

Relatively little is known about the distribution of the ^{137}Cs isotope in various elements of the environment of Poland and other European countries prior to the Chernobyl disaster. Considering the half-life of ^{137}Cs and the time that has elapsed since the nuclear testing in Europe, determining the activity of the isotope in stands representing different age groups seemed to be an interesting study objective. Apart from the analysis of the activity of the artificial ^{137}Cs isotope, the aim of the research was the analysis of the activity of the natural ^{40}K isotope. Two tree species, *Pinus sylvestris* and *Betula pendula*, occurring in a selected forest district in eastern Poland, were chosen as the research object. The activity of ^{137}Cs and ^{40}K isotopes was measured in three stands of different ages, approximately 30, 50, and 80 years old. The following research hypotheses were adopted: (i) the species and age of the tree determines the activity of ^{137}Cs and ^{40}K ; (ii) both the species and age of the stand influence the values of the coefficients transfer of ^{137}Cs and ^{40}K ; and (iii) the activity of ^{137}Cs in stands of different ages enables the assessment of radioactive contamination of the studied area in the past.

2. Materials and Methods

The research area was located in eastern Poland (Sokołów Forest District; a longitude of $21^{\circ}54' - 22^{\circ}33'$ E and a latitude of $52^{\circ}17' - 52^{\circ}42'$ N). The area of forestry is approximately 2500 ha. The dominant species in the stand is pine (roughly 50% of the area). The percentage of deciduous trees is much lower, with birch covering approximately 15% of the forested area. The age structure of stands is dominated by trees over 60 years old, occupying approximately 30% of the area of the forest district (information obtained from the Sokołów Forest District).

A total of 36 trees (6 individual trees of each species and age group) were selected for the study, representing two species of Scots pine *P. sylvestris* and silver birch *B. pendula* and belonging to three age groups: 30, 50, and 80 years old. Samples for testing were collected during the logging period carried out in the Forest District, i.e., in October–November 2017. Three to seven rings were cut from each tree (the total weight of the sample collected

from each tree was at least 10 kg). The rings (including bark) were weighed, then dried for one year at room temperature and incinerated. The ash obtained from each sample was weighed and subjected to γ -spectrometric analysis.

Concurrently, litter and soil samples were collected from the sites of the wood sampling. The litter was gathered from an area of 2×2 m, and soil samples were obtained from a depth of about 30 cm. Litter and soil samples were air dried in the laboratory. Then, the soil was sieved through a sieve with a mesh diameter of 2 mm. As with the ash, the litter and soil samples were subjected to γ -spectrometric analysis.

Gamma-spectrometric analysis was performed with a Canberra spectrometer. Spectrum analyses were carried out using the Genie-2000 software package (Gamma Analysis Option model S 5001 C). Each sample was measured for 180,000 s. The detection limits were 0.15 Bq/kg for ^{137}Cs and 2.5 Bq/kg for ^{40}K . The results of the measurements of the activity of ^{137}Cs and ^{40}K in the ash obtained from wood were converted into the activity of isotopes in fresh weight of 1 Mg (tons) of wood.

Using the activity of ^{137}Cs and ^{40}K isotopes in wood per 1 kg and the activity of these isotopes in soil, the values of transfer coefficients (TF) of ^{137}Cs and ^{40}K isotopes were calculated as the ratio of their activity in fresh wood mass (Bq/kg) to their activity in soil (Bq/kg).

Statistical analyses were performed with *Statistica* software v. 13 (TIBCO Software Inc., Palo Alto, CA, USA). The level of significance was 0.05. The differences in isotope activity in wood, litter, and soil were tested by two-way ANOVA (tree species and age of the stands as two factors). Due to the heterogeneity of the variance in the tested groups of variables, the data were logarithmized. Multiple comparisons between groups of variables were performed by the Tukey test. The differences in TF values for the age groups of trees were tested with the nonparametric Kruskal–Wallis ANOVA and post hoc Dunn test. Differences in TF between pine and birch for the same tree age were tested with Mann–Whitney test. The influence of potassium content in soil on the value of the TF ^{137}Cs was determined using the Sparman's rank correlation coefficient.

3. Results

The average activity of ^{137}Cs in the studied tree species ranged from 150.3 Bq/Mg (30 year-old pine) to 3696 Bq/Mg (80 year-old birch). The activity of ^{137}Cs in wood depended on the tree species and the age of the stand. Higher levels of activity of ^{137}Cs were recorded in birch wood compared to pine wood. The highest activity levels of the isotope were recorded in the oldest wood ($p = 0.0001$) (Table 1, Figure 1A).

The mean activity of ^{137}Cs in the litter ranged from 12.72 Bq/kg to 99.05 Bq/kg. The activity of the isotope increased with age only in the birch stand ($p < 0.05$) (Table 1, Figure 1B).

The cesium isotope showed similar levels of activity in all soil samples, regardless of the age and the type of the stand. The average activity levels did not exceed 27 Bq/kg (Table 1, Figure 1C).

The average activity of ^{40}K in the litter ranged from 21.3 to 156.5 Bq/kg. Potassium accumulation was determined by the type of stand and its age. The litter in the birch stand was characterized by a higher potassium activity compared to the litter in the pine stand. The lowest potassium activity was recorded in the litter of the youngest stand ($p < 0.001$) (Table 2, Figure 2B).

The average ^{40}K activity in the soil ranged from 126.7 Bq/kg for an 80 year-old birch stand to 328.1 Bq/kg for a 30 year-old birch stand. The highest activity of ^{40}K was recorded in the soil of the youngest stands. The activity of ^{40}K decreased with the age of the stand, and the tree species had no significant influence on its activity in the soil (Table 2, Figure 2C).

Table 1. Results of two-way ANOVA showing the impact of two factors (two tree species and three tree ages) on ^{137}Cs activity in wood (A), litter (B), and soil (C) of *Pinus sylvestris* and *Betula pendula* stands. The sum of squares (SS), degrees of freedom (df), mean of squares (MS), *F*-ratio (*F*), and probability value (*p*-value) are shown; statistically significant results are in bold.

Parameters	SS	df	MS	<i>F</i>	<i>p</i> -Value
WOOD					
Species	4.074	1	4.074	20.93	0.0001
Age	54.07	2	27.04	138.9	<0.0001
Species $\times$ age	0.294	2	0.147	0.754	0.4790
Error	5.839	30	0.195		
LITTER					
Species	0.006	1	0.006	0.026	0.8740
Age	10.01	2	5.006	23.46	<0.0001
Species $\times$ age	3.216	2	1.608	7.536	0.0022
Error	6.402	30	0.213		
SOIL					
Species	12.24	1	12.24	0.455	0.5051
Age	132.5	2	66.24	2.463	0.1022
Species $\times$ age	59.59	2	29.80	1.108	0.3434
Error	806.8	30	26.89		

Table 2. Results of two-way ANOVA showing the impact of two factors (two tree species and three tree ages) on ^{40}K activity in wood (A), in the litter (B), and in the soil (C) of *Pinus sylvestris* and *Betula pendula* stands. Sum of squares (SS), degrees of freedom (df), mean of squares (MS), *F*-ratio (*F*) and probability value (*p*-value) are shown; statistically significant results are in bold.

Parameters	SS	df	MS	<i>F</i>	<i>p</i> -Value
WOOD					
Species	1.510	1	1.510	34.89	<0.0001
Age	2.302	2	1.151	26.59	<0.0001
Species $\times$ age	0.129	2	0.064	1.489	0.2418
Error	1.299	30	0.043		
LITTER					
Species	3.936	1	3.936	10.85	0.0025
Age	8.002	2	4.001	11.03	0.0003
Species $\times$ age	0.710	2	0.355	0.978	0.3877
Error	10.88	30	0.363		
SOIL					
Species	7485	1	7485	1.684	0.2043
Age	210,760	2	105,380	23.70	<0.0001
Species $\times$ age	4866	2	2432	0.547	0.5842
Error	133,370	30	4446

Figure 1. Activity of ^{137}Cs in wood (A), litter (B), and soil (C) in pine (P) and birch (B) stands of different ages (mean $\pm$ confidence level). Different letters denote statistically significant differences among means in Tukey test ($p < 0.05$) after two-way ANOVA for the interaction between species and age (see Table 1).

The ^{137}Cs transfer coefficient had the highest values for the oldest trees ($p < 0.001$). The highest values of $\text{TF } ^{137}\text{Cs}$ were recorded for 80 year-old birch, and they were significantly higher than those noted for pine ($p = 0.0453$) (Figure 3A). There was a significant correlation between the activity of ^{40}K in soil and the $\text{TF } ^{137}\text{Cs}$ value ($r_s = -0.540$, $p < 0.001$). The age of the trees did not affect the $\text{TF } ^{40}\text{K}$ value of any of the species, while significant differences in TF were noted for birch, compared to pine, in the 30 and 50 year-old stands ($p < 0.05$) (Figure 3B).

Figure 2. Activity of ^{40}K in wood (A), litter (B), and soil (C) in pine (P) and birch (B) stands of different ages (mean $\pm$ confidence level).

Figure 3. Values of ^{137}Cs (A) and ^{40}K (B) transfer coefficients (TF) in pine (P) and birch (B) stands of different ages (median $\pm$ quartile). Different letters denote statistically significant differences in Dunn test after Kruskal–Wallis ANOVA or between tree species inside each age group in Mann–Whitney test ($p < 0.05$).

4. Discussion

Our research has demonstrated that, even several decades after the radioactive contamination of the environment, the ^{137}Cs isotope is still retained in forests in soil, litter, and wood. The greatest activity of ^{137}Cs is observed in the surface layer of the soil [9,12], mainly in the litter. This observation applies to ^{137}Cs released into the environment both after the Chernobyl accident and during the 1950s and 1960s [17,18]. Radiocesium is recycled in forest ecosystems [19].

The activity of radioactive isotopes in plants depends on many factors, including the scale of radioactive contamination, tree species, isotopic composition of pollutants, and agrochemical properties of soil and time elapsed since the isotope's release [3,13]. In our research, the highest activity of the ^{137}Cs isotope was recorded in the oldest trees, the growth period of which coincided with the era of intensive nuclear weapons testing. The activity of cesium in the wood of the oldest stands was over 10 times higher than the activity measured in the wood of 50 and 30 year-old trees. Taking into account the half-life of the ^{137}Cs isotope, the time that has elapsed since the period of nuclear weapons testing and the Chernobyl disaster, as well as the currently measured activity of ^{137}Cs in the wood of stands of various age groups, it was found that the nuclear weapons testing conducted for several years during the 1950s and 1960s led to even greater (up to 30 times more) environmental contamination in eastern Poland than the Chernobyl disaster in 1986. The results of our research confirm earlier reports from northern Europe [20–22].

Our study chose not to focus on the Fukushima reactor accident in 2011 because the activity of radioactive elements emitted during the Fukushima accident in Japan was approximately four orders of magnitude lower than during the explosion of the Chernobyl nuclear reactor [23]. The deposition of radionuclides after the Fukushima accident did not affect the environment in Poland. For comparison, the Cs radioactivity in the air measured in Warsaw on 28–30 April 1986 was $6570 \text{ mBq } ^{137}\text{Cs}/\text{m}^3$, and on 28–30 March 2011, it was $0.34 \text{ mBq } ^{137}\text{Cs}/\text{m}^3$ [24].

The contamination of trees with ^{137}Cs is the result of different processes such as uptake by the root system, internal translocation and immobilization [25]. Cesium is strongly accumulated by tree roots, especially by birch roots [8,26]. The differences in the greater activity of cesium in birch wood than in pine can be explained by the fact that deciduous trees generally show a greater tendency to branching roots than conifers [27,28], which results in a larger surface for uptake of substances from the soil. The differences in the absorption of ^{137}Cs by trees are also confirmed by the studies of Immamura et al. [29], carried out on oak and pine in Japan after the Fukushima accident. The authors noted that oak trees exhibited stronger ^{137}Cs absorption capacities than pine trees.

In the studies conducted eight years after the Chernobyl accident in Lithuania [30], higher radioactivity levels of ^{137}Cs were recorded in pine forest litter than in birch forest litter. In our research, we did not find any differences in the ^{137}Cs activity in the litter of two types of stands. However, we observed an increase in the activity of this isotope with regard to the age of the birch stand. The assimilation organs (leaves, needles) reveal the highest activity of ^{137}Cs compared to other parts of the tree [31,32]. Our research indicated that the activity of ^{137}Cs (Bq/kg) in the studied components occurred in the following order: litter > soil > wood. The sources of cesium in the litter are falling assimilation organs and parts of trees covered with bark (branches, trunk bark). In the latter, the activity of cesium increases with age [33], and it is especially visible in birch stands [34,35]. In the case of potassium, the litter in 50 and 80 year-old birch stands accumulated more potassium than in pine stands, which is consistent with other literature reports [36,37]. On the other hand, the soil was characterized by the highest activity of ^{40}K in the youngest stands.

These results may indicate a change in land use—the youngest stands were probably planted on previously used agricultural soils with a high potassium content. It should also be noted that the oldest stands with the highest activity levels of ^{137}Cs were characterized by the lowest activity levels of ^{40}K . Cesium enters plants mainly through the potassium transport systems [38]. Increasing the potassium content in the soil reduces the uptake of ^{137}Cs by plants [12,13]. Our research showed that the higher the potassium content in the soil, the lower the ^{137}Cs translocation coefficient. Similar results were reported, for example, by Duong et al. [39], who studied ^{137}Cs and ^{40}K activity in *Acacia auriculiformis* trees, and by Guillaume et al. [40], who assessed the translocation of ^{137}Cs in alpine plants.

Relatively high TF values for ^{137}Cs in 80-year-old birch wood are also noteworthy. As mentioned previously, the oldest birch stands accumulate more ^{137}Cs than pine stands [34,35]. This is probably due to the immobilization of this isotope, e.g., in the bark of trees. According to the data cited by Bojko [4], in the forests of the northern regions of the Zhytomyr region (Ukraine), the activity of ^{137}Cs in un-debarked wood ranged from 80 to 1400 Bq/kg, and in barkless wood, it was slightly lower and amounted to 50–1100 Bq/kg. Similarly, in the Kiev region (Ukraine), the activity was 70–820 Bq/kg and 45–300 Bq/kg, respectively. In trees, old and perennial tissues and organs are the most contaminated, e.g., the outer bark of trees, which is directly exposed to radioactive fallout [33,41,42]. The concentration of Cs in the bark of trees is largely determined by the direct adsorption of the radionuclide on the tree surface after the occurrence of radiocontamination [41,42]. Research carried out in Japan six months after the nuclear reactor accident in Fukushima [42] showed that greater radiocesium activity was recorded in the bark than in the inner parts of the trunk (sapwood and heartwood), and the ^{137}Cs activity in oak bark was greater than in pine bark. The bark of deciduous trees is more stable than that of conifers [43,44], which makes it peel to a lesser extent. In our research, we determined the activity of isotopes in un-debarked wood.

It can be assumed that the greater accumulation of ^{137}Cs in birch wood than in pine trees in the oldest stands noted in our study is due to the greater activity of this isotope in birch bark than in pine bark. The bark of trees is a good bioindicator of air pollutants, including heavy metals [45–49]. Taking into account the obtained data, it can be concluded that stands over 75 years of age can be used to assess radioactive contamination of the

environment in the past; due to the greater activity of ^{137}Cs , birch stands seem to be more useful for bioindication purposes than pine stands.

The higher values of the potassium transfer coefficient recorded in deciduous stands compared to coniferous stands resulted from the faster mobilization of this element in deciduous forests. Deciduous stands have a much shorter residence time of basic cations, e.g., K in soil organic layers compared to coniferous tree habitats [50]. This is probably due to the faster decomposition of the litter of deciduous forests than coniferous ones [51]. The decomposition rate of organic matter in coniferous forests is approximately 17 years, and in deciduous forests, it is approximately four years [52].

5. Conclusions

The analysis of the activity of the ^{137}Cs isotope in pine and birch stands of different ages in a selected area of eastern Poland made it possible to assess the radioactive contamination of the environment over several decades.

The results of measurements of ^{137}Cs activity in 80 year-old stands showed that the radioactive contamination of the environment in the studied region of Poland during the nuclear weapons testing during the 1950s and 1960s was up to 30 times higher than in the period after the Chernobyl disaster.

Greater levels of ^{137}Cs and ^{40}K accumulate in birch than in pine. This was also confirmed by the values of transfer coefficients: higher values of transfer coefficients were shown by the ^{137}Cs isotope in birch wood than in pine wood, especially with regard to the oldest stands.

As the age of the stand increases, the accumulation of ^{137}Cs in wood also increases, while the accumulation of ^{40}K decreases.

The higher activity of ^{137}Cs found in birch wood indicates a greater usefulness of deciduous trees when assessing radioactive contamination of the environment.

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Data Availability Statement: The data presented in this study are available on request from the corresponding author.

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