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Accumulation and Distribution of ^{137}Cs and ^{90}Sr Radionuclides in the Forests of the «Drevlyansky» Nature Reserve

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Abstract: This study was conducted in order to define the main physical and chemical properties of the soil, data on the accumulation as well as transfer of ^{137}Cs and ^{90}Sr radionuclides in the main components of the forest biocenosis of the Drevlyansky Nature Reserve, which is located in the area affected by radiation from the Chernobyl nuclear power plant catastrophy. The processes of radionuclide migration by the soil profile have been studied, the highest level of pollution density in the upper soil layers has been noted in the layers of 0-2, 2-4 cm. The analysis of the main agrophysical indicators of soils is carried out in the main forest types of for the territory of the Drevlyansky Nature Reserve. The process of radionuclides migration from soil to plants was also studied by determining the coefficient of soil to plant transfer factor (TF) and accumulation (A) of such radionuclides as ^{137}Cs and ^{90}Sr. The maximum transition coefficient was recorded during migration to *Calluna vulgaris (L.) Hill.*, – 33,38 for ^{137}Cs and 34,97 for ^{90}Sr and the lowest to *Vaccinium vitis-idaea L.* – 2,38 for ^{137}Cs and 9,27 for ^{90}Sr An increase in the transition coefficient was observed with increasing of soil moisture.

Keywords: Natural Reserve, Radionuclides, Transfer Factor, Activity Concentration

1. Introduction

Already in the first days after the Chernobyl accident, one of the largest anthropogenic disasters of mankind, researchers engaged in radioecological survey of areas, noted higher levels of radioactive contamination of forests compared to other forms of landscapes [1-3]. It was found that forest lands contained 20–30% more radioactive elements compared to the open area [4-6].

A critical situation was noted in the forests of Narodychi district, where the area of forests with a density of radioactive contamination of the soil was over 555 kBq/m^2 on 14.9 thousand hectares and on the area of 4.5 thousand hectares over 1480 kBq/m^2. More than 35 years have passed since the Chernobyl accident, the radiation situation has changed, but the main holders of radionuclides are still forest floor and soil, which continue to affect the rate and direction of their migration to other components of ecosystems and food chains to humans. In particular, it has been established that forest floor is the main depot of radioactive elements, in which the content of specific activity of ^{137}Cs and ^{90}Sr is 100 or even 1000 times higher than in other components of forest ecosystems. [3, 5, 7] It's especially relative to pine forests where total deposition of the ^{137}Cs in forest floor exceeding 40% of the total deposition in the pine forest ecosystem [8] The researchers also found that the ability of forest floor to hold ^{137}Cs and ^{90}Sr depends on its capacity, structure, composition, age of plantations and grass cover, and the presence or absence of moss cover. It may also change with time due to ecological processes, such as differences in the root absorption rate and changes in the allocation pattern [9]

At the same time, it was found that in pine plantations a strong layer of forest floor is formed, which increases the holding capacity of radionuclides more than in deciduous ones. Slow mineralization of coniferous-moss precipitation (6–10 years) causes migration of a small part of radioactive elements to the humus-eluvial horizon, and in deciduous and mixed forest stands this movement occurs at a faster rate [4, 9]. Therefore, the content of radionuclides in the mineral layers of the soil is directly dependent on the concentration of radioactive elements in the forest floor.

Determinants of migration of radionuclides to plants are physicochemical parameters of the soil and the systematic affiliation of plants. The latter is of great practical importance, as it allows to assess their qualitative level of possible radioactive contamination due to the systematic situation of certain species,. With the exception of radiocaesium, the intensity of radionuclide accumulation by plants from the soil has been studied only in fragments.

More than 55% of the territory of the Drevlyansky Nature Reserve is occupied by forests. Among them, about 50% are areas with the type of forest vegetation conditions – Bir. Bir is a type of forest vegetation conditions characterized by very poor soils, oligotrophic vegetation according to Pogrebnyak's typology of the forest. There are 6 types of bir according to the level of soil moisture: very dry (A0); dry (A1); fresh (A2); humid (A3); wet (A4); waterlogged (A5) in Pogrebnyak's typology [10]. That is why it is necessary to study the current levels of radioactive contamination of various components of forest ecosystems, in particular – on the vertical migration of radionuclides in forest soils. Implementation of a set of studies to study the migration of ^{137}Cs and ^{90}Sr and assess the state of forest ecosystems with the type of forest vegetation conditions bir, which will optimize the management of nature reserves in the radioactively contaminated areas.

2. Materials and Methods

2.1. Study Area

Drevlyansky Nature Reserve (hereinafter the reserve) is located in Zhytomyr region, Narodytsky district to the south and southeast of the village.

The reserve was established on December 11, 2009 on the area of 30872.84 hectares. The territory of the reserve is characterized by the composition of biocenoses typical for the Polissya region of Ukraine and the absence of anthropogenic pressure.

2.2. Soil and Plant Sampling

Sampling took place on the territory of the reserve in forest areas with the type of forest vegetation conditions – bir, during June – August 2019 and 2020, on the territory of Moteykivsky and Narodytsky departments of the reserve. The predominant soil types are represented by sod-podzolic and sod-strongly podzolic soils.

Sampling of soil at a depth of 0-20 cm, to determine ^{137}Cs, ^{90}Sr was carried out according to State standarts of Ukraine (hereinafter DSTU) (4287: 2004 "Soil quality. Sampling "and according to the" Methodology of comprehensive radiation survey of contaminated areas as a result of the Chornobyl disaster (except for the territory of the exclusion zone)". Sampling of plant origin for the determination of ^{90}Sr and ^{137}Cs was carried out according to the guidelines "Sampling, primary processing and determination of the content of ^{90}Sr and ^{137}Cs in plant products." Sampling was carried out at least 50 m from roads, any objects of human activity and from areas with a sharply different from the background state of vegetation. The general sample consisted of 20 spot samples taken near the attachment point.

Samples of plant phytomass were taken on the territory of Narodytsky department of the reserve on 12 plots with type of forest conditions A2, and on the territory of Moteykivka department on 12 plots with type of forest conditions A1 and 12 plots with type of forest conditions A3. Chosen species represent a few of the most common plants in these forest types [11].

2.3. Determination of Activity Concentrations

Preparation and determination of ^{137}Cs and ^{90}Sr concentration in soil, plant phytomass and forest floor was performed by spectrometric method using a scintillation gamma-betta spectrometer MKS-AT 1315 according to DSTU 7868: 2015 and DSTU 7867: 2015.

2.4. Determination of Transfer Factors (TFs)

The coefficient of transition of radionuclides from soil to plants (hereinafter TF) is calculated as the ratio of the specific activity of dry matter of the plant to the density of soil contamination by radionuclide:

$$\mathrm{TF} = \frac{Am}{Si}$$

where:

A_m – specific activity of the radionuclide dry matter of the plant, Bq/kg;

S_i — density of soil contamination by a certain radionuclide, kBq/m^2,

The coefficient of accumulation (hereinafter A) is calculated as the ratio of the specific activity of the radionuclide in the plant to its specific activity in the soil.

$$A = \frac{Am}{As}$$

Where: A_m – specific activity of the radionuclide dry matter of the plant, Bq/kg; A_s — specific activity of soil radionuclide by a certain radionuclide, kBq/m^2.

3. Results

3.1. Agrochemical Soil Properties

One of the determining factors of the radionuclide migration nature is the agrochemical properties of the soil. Table 1 shows the generalized data of the analysis of soil samples taken from the main types of forest vegetation conditions, namely: dry (A1), fresh (A2) and humid (A3).

In A1 samples the soil is characterized by a very strongly acidic reaction of the environment, a very low amount of absorbed bases, a very low content of humus, easily hydrolyzed nitrogen compounds, exchangeable potassium, calcium, sulfur. The soil sample in A2 has a moderately acidic reaction, a very low amount of absorbed bases, low content of humus, easily hydrolyzed nitrogen, exchangeable potassium, calcium, sulfur, medium mobile phosphorus. In A3 samples, the soil has a neutral reaction, a very low amount of absorbed bases, low content of humus, easily hydrolyzed nitrogen, exchangeable potassium, calcium, low sulfur content and high mobile phosphorus.

Table 1. *Agrochemical properties of the soil.*

Characteristic	A1	A2	A3
Metabolic acidity, pH	3,28±0,35	4,91±0,53	6,6±0,91
Hydrolytic acidity, mmol/100 g of soil	5,8±0,51	3,12±0,30	0,99±0,11
The sum of absorbed bases, meq/100 g of soil	3,2±0,32	1,4±0,16	3,0±0,34
Humus,%	0,84±0,096	0,93±0,12	1,24±0,14
Nitrogen-hydrolyzed nitrogen, mg/kg of soil	68,6±7,88	28,0±4,4	22,4±3,11
Mobile phosphorus, mg/kg of soil	34,05±4,7	55,6±7,72	131,9±20,29
Exchangeable potassium, mg/kg of soil	20,8±2,41	15,8±1,81	33,6±4,6
Calcium, mmol/100 g of soil	1,50±0,16	0,87±0,09	1,0±0,11
Sulfur, mmol/100 g of soil	2,35±0,25	4,43±0,59	5,13±0,58
Copper	0,13±0,01	0,20±0,02	0,47±0,05
Zinc	1,73±0,18	1,67±0,22	0,85±0,09
Cobalt	0,113±0.01	0,109±0.01	< 0,085±0.01
Cadmium	0,022±0,002	0,031±0,002	0,015±0,001
Lead	1,65±0,17	1,8±0,21	1,66±0,19
Manganese	12,33±1,41	14,61±2,02	20,40±2,34

Exceedances of maximum permissible concentrations were not detected in the content of heavy metals in all studied types of forest.

3.2. ^{137}Cs and ^{90}Sr Pollution Density in Soil

The data obtained as a result of layer-by-layer analysis of the specific activity of ^{137}Cs in the soil is expressed in table 2. The peak indicator of pollution density was recorded in the soil layer at a depth of 0-2 and 2-4 cm in all studied types of forest. In A1 the share of radionuclides in the soil layer 0-4 cm is 68.71% in A2 65.69% and in A3 65.82% of the total (Table 3) With increasing depth, the density of pollution decreased. In particular, the largest difference is the decrease in the density of contamination in the layer of 4-6 cm in comparison to the layer of 2-4 cm in 1.3 times for A1 and 1.4 for A2 and A3, and in the layer of 8-10 cm in comparison to 6-8 cm 2.7 times in A1, 1.8 in A2 and 1.7 in A3. However, there is a slight increase from a layer of 12-14 cm to 14-16 cm by 13.5% and from a layer of 16-18 cm to 18-20 cm by 29% in A1 and A2 from 14-16 to 16-18 at 5.8%.

Table 2. *Layer-by-layer analysis of pollution density (kBq / m^2).*

Depth of sampling, см	A1		A2		A3	
	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr
0-2	504,2±112,2	7,5±2,0	621,7±60,9	9,2±1,08	548,1±56,4	8,4±0,91
2-4	425,6±78,5	8,2±2,2	582,3±63,3	9±0,94	527,7±54,3	8,1±0,83
4-6	182,4±30,5	3,7±0,8	241,1±27,7	9,7±0,99	218,1±21,4	6,9±0,71
6-8	125,1±21,1	9,4±2,7	191,8±20,3	7,8±0,80	172,8±18,8	6,7±0,77
8-10	33,4±5,2	5,2±1,9	68,8±7,3	4,2±0,44	63,4±7,3	5,6±0,59
10-12	18,4±2,9	3,6±0,6	31,6±3,3	3,9±0,38	28,9±2,5	3,1±0,30
12-14	13,3±2,8	3,4±0,4	27,8±2,9	4,2±0,44	21,3±2,1	3±0,29
14-16	15,1±3,5	1,8±0,3	24,42±2,5	3,2±0,33	20,7±2,2	3,2±0,36
16-18	14,9±3,2	2,9±0,4	25,9±2,9	3,1±0,31	17,9±1,8	2,2±0,23
18-20	20,8±3,8	4,3±0,4	17,2±1,7	2,9±0,33	15,6±1,7	1,9±0,20

The data obtained as a result of layer-by-layer analysis of the specific activity of ^{90}Sr in the soil indicate an uneven distribution of contamination density of the soil profile in the type of forest vegetation A1, so the peak value of 9.4±2.7 kBq/m^2 the maximum difference is recorded between the layers of 2-4 and 4-6 cm, a decrease of 1.2 times and between 4-6 and 6-8 cm, an increase of 1.5 times. Determining the reasons for the diversity of indicators of the specific activity of this radionuclide requires further research. Indicators of A2 pollution density decrease with increasing depth except for layers 4-6; and 12-14 cm. The peak index of 9.7±0.99 kBq/m^2 was recorded in the 4-6 cm layer. In A3, the peak index was recorded in the 0-2 cm layer. and is 8.4±0.91 kBq/m^2, there is also a decrease in the proportion of radionuclides with increasing depth except for the layer 14-16 cm, the density of which is 20% higher than in the layer 12-14 cm.

A regression analysis of the dependence of the increase in the proportion of radionuclides in the lower soil layers on the type of forest growing conditions was also performed.

Calculations showed that the correlation coefficient for ^{137}Cs is 0.687, the significance coefficients are close to zero (0.002), which indicates a reliability of the relationship at 68.7%. Thus, the obtained dependences are close and the equations are reliable.

However, the correlation coefficient for ^{90}Sr is less than 0.5, namely 0.39, which indicates the inaccuracy of the dependence of the migration of radionuclides down the soil profile on the type of forest vegetation conditions.

Within the types of habitat conditions, it is advisable to group plants according to the intensity of their accumulation of a certain radionuclide, in particular, ^{137}Cs.

Table 3. *^{137}Cs accumulation parameters in plant phytomass.*

Forest vegetation conditions	Scientific specie name	Pollution density kBq/m^2	Activity concentrations Bq / kg		A_{137Cs}	TF, м^2 kg^{-1} 10^{-3}
			plant phytomass	Soil		
A1	Calluna vulgaris (L.) Hill. (Aboveground part)	66,5±7,6	1415±149	335,4±36,5	4,22	21,26
	Vaccinium vitis-idaea L.	1848,2±200,8	4400±464	9314,9±1012,5	0,47	2,38
A2	Calamagrostis epigeios (L.) Roth	127,2±14,6	1587±164	640,9±69,6	2,47	12,48
	Calluna vulgaris (L.) Hill.	252,8±27,5	8442±869	1274,2±131,2	6,62	33,38
A3	Vaccinium vitis-idaea L.	293,5±30,9	1338±145	1479,4±154,1	0,90	4,55
	Vaccinium myrtillus L. (Aboveground part)	180,8±17,7	2943±319	911,2±99,1	3,23	16,28

3.3. Characteristic of Soil to Plant Radionuclides Transfer

Given the value of TF, the intensity of ^{137}Cs to the phytomass is divided into very strong (TF > 100), strong (100> TF > 50), moderate (50> TF > 10), weak (10> TF > 1), very weak TF <1) [13, 14]. The results of the analysis of the transition of radionuclides into plants typical of Polissya forests are shown in table 3.

As a result of research, it was proved that the plant samples are in the range of moderate and low intensity of ^{137}Cs to phytomass, namely: *Calluna vulgaris, Calamagrostis epigeios (L.) Roth* and *Vaccinium myrtillus L.* have a moderate intensity, while *Vaccinium vitis- idaea L.* has a low intensity. Among the studied samples, the highest coefficient of transition to soil has *Calluna vulgaris (L.) Hill.* – 33.38 in more humid conditions of A2, and the lowest indicator is discovered in *Vaccinium vitis-idaea L.* samples in A1 2.38.

Table 4. *^{90}Sr accumulation parameters in plant phytomass.*

Forest vegetation conditions	Scientific specie name	Pollution density kBq/m^2	Activity concentrations Bq/kg		A_{90Sr}	TF, м^2 kg^{-1} 10^{-3}
			plant phytomass	soil		
A1	Calluna vulgaris (L.) Hill. (Aboveground part)	1,48±0,16	26,6±3,69	7,4±0,81	3,59	17,97
	Vaccinium vitis-idaea L.	2,07±0,23	19,2±2,2	10,36±1,19	1,85	9,27
A2	Calamagrostis epigeios (L.) Roth	2,59±0,29	70,1±8,73	30,21±3,47	2,32	27,06
	Calluna vulgaris (L.) Hill.	1,55±0,17	54,2±6,08	7,82±0,83	6,93	34,97
A3	Vaccinium vitis-idaea L.	2,14±0,33	28,3±3,25	10,7±1,14	2,64	13,22
	Vaccinium myrtillus L. (Aboveground part)	53,81±6,28	1670±190,86	545±62,6	3,06	31,05

The maximum and minimum values of the transition coefficient ^{90}Sr were also recorded in samples of *Calluna vulgaris (L.) Hill.* – 34.97 in A2, and the lowest rate was found in *Vaccinium vitis-idaea L.* samples in A1 – 9.27. ^{90}Sr transition coefficients for *Calamagrostis epigeios (L.) Roth* and *Vaccinium myrtillus L.* are higher than ^{137}Cs, namely 116% and 90%, respectively.

4. Conclusion

The obtained results and conclusions correspond to the results of researches of other scientists and confirm their relevance for the territory of Drevlyansky reserve [15, 16]. The highest indicators of pollution density are found in the upper layer of the soil (0-4 cm). With a sufficient level of reliability, it is proved that with the change of the type of forest vegetation conditions from A1 to A2 and A3, the bulk of ^{137}Cs radionuclides migrates down the soil profile.

This is the first study of TF and A^{137}Cs, A^{90}Sr determination for forest plants in the Chernobyl catastrophe remote period in Ukraine. The studied plant samples are characterized by moderate and low intensity of ^{137}Cs migration to phytomass. And increased in comparison with ^{137}Cs transition coefficients for ^{90}Sr. There is an increase in the coefficients of accumulation and transition for the type of forest vegetation conditions which are defined with a higher level of soil moisture.

This work not only examines the current state of movement of radionuclides in the ecosystem in the radioactively contaminated area of the nature reserve, but also expands the database of coefficients of transition of radionuclides into plants characteristic of forest ecosystems. In order to fulfil blanks in the information about territories with other types of forest vegetation and their interaction investigation will be continued in the future.

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