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## ARTIFICIAL RADIONUCLIDES IN THE FOREST LITTER AT THE TERRITORY ADJACENT TO SEMIPALATINSK TEST SITE

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The paper provides results of research of concentration and distribution of artificial radionuclides in the components of forest litter and in the top soil layer of the pine forest of Priirtyshie long after nuclear tests at the territory of Semipalatinsk Test Site (the STS). Minimum and maximum values of specific activity were determined for  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$ ,  $^{239+240}\text{Pu}$  in the soils researched as well as in mineral and organic components of litter. Series of radionuclides were formed depending on their ability to be accumulated in the top layer of soil and in the components of forest litter. Dependence between concentrations of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$ ,  $^{239+240}\text{Pu}$  in the litter and its thickness was determined.

**Keywords:** radioecology, Semipalatinsk Test Site (the STS), artificial radionuclides ( $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$ ,  $^{239+240}\text{Pu}$ ), forest ecosystem, forest litter.

### INTRODUCTION

Semipalatinsk Test Site (STS) – is one of the largest test sites used for testing nuclear weapons in the world. It occupies the area of 18 300 km<sup>2</sup>. For 42 years of operation, 456 nuclear tests using 616 devices were conducted here. The tests conducted at the STS caused contamination not only of the test site territory itself but also, beyond its boundaries. So, as the result of the test of the 29<sup>th</sup> of August, 1949, territory of pine forest of Priirtyshie suffered contamination (Figure 1).

Processes of radionuclides migration in forest biocenoses have a series of peculiarities comparing with other natural plant communities, due to large area of above-ground phytomass, multi-year cycle of plants development, availability of forest litter and etc. [1].

The main primary absorbers of radionuclides in forest ecosystems include leafage and needles, forest undergrowth, as well as forest litter. Radioactive substances can be flushed off with precipitations, get on soil surface

when leaves fall from the plants, and also, when stalks (of grass, for example) die off. Only moss and lichens can retain radionuclides indefinitely. Therefore, irrespective of season, when the fallout took place, major part of contamination proves to be in the litter [2, 3].

Forest litter is the first soil layer, where radionuclides get accumulated, transformed, and from where their migration starts. In course of time, radionuclides entering the soil litter and top layers of soil get redistributed in such a manner that they enrich the soil layers below with mobile compounds. At that, decomposing forest litter serves as a potential source of mobile forms of radionuclides that enter soil [4, 5].

The aim of this research is to study composition and distribution of artificial radionuclides ( $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$ ,  $^{239+240}\text{Pu}$ ) in forest litter and in the top soil layer of pine forest of Priirtyshie long after the nuclear tests at the STS territory.

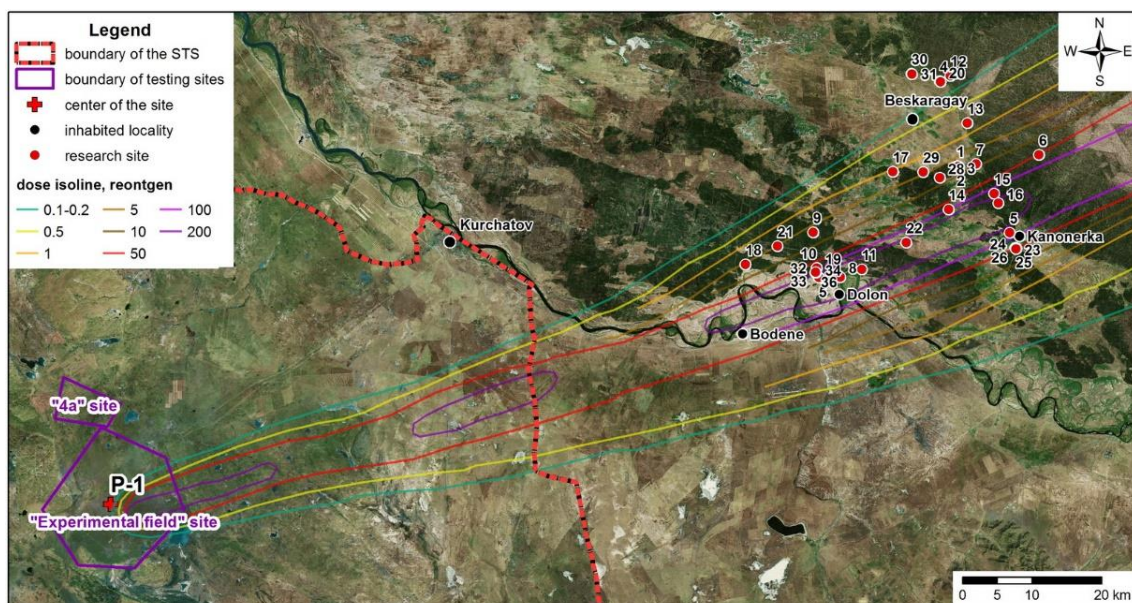


Figure 1. Study area

### MATERIALS AND METHODS

As the research areas, forest plots within the plume of the 1949, with presumably increased concentrations of radionuclides in the environmental components were chosen. In total 36 research areas were arranged (Figure 1). From each site, samples of litter and mixed samples of soil were taken to the depth of 5cm using envelope method.

Litter was dried under natural conditions. After that, to separate organic components from mineral components, the sample was sieved through a 2 mm mesh. Organic component was charred until black residue, milled using laboratory mill to reach homogenous mass. After that, specimens were placed into crucibles for further ashing. Initial temperature was increased to 200 °C for 50–60 minutes, after that the maximum temperature of muffle furnace was set as follows: ashing temperature for further determination of <sup>137</sup>Cs is 400 °C, while for <sup>90</sup>Sr, <sup>241</sup>Am and <sup>239+240</sup>Pu it's up to 550 °C.

Samples were dried to the air-dry state in drying cabinets at the temperature of 50–60 °C. After coarse stones and inclusions (plant residues) were removed, dried samples were weighed using counterbalance. Then the entire soil mass was carefully mixed, ground in a mortar of porcelain using pestle and sieved using a 1 mm mesh. To check whether the whole amount of sample was sieved, or not, each sieve was shaken above a sheet of paper.

Analyses to measure activity concentrations of radionuclides in samples were conducted in accordance to GOST guidelines with a certified laboratory equipment [6–7]. Activity concentrations of <sup>137</sup>Cs and <sup>241</sup>Am were determined with a  $\gamma$ -spectrometer *Canberra GX-2020*, <sup>90</sup>Sr and <sup>239+240</sup>Pu were determined using a radiochemical isolation followed by measuring with a  $\beta$ -spectrometer *TRI-CARB 2900 TR* and  $\alpha$ -spectrometer *Canberra (mod. 7401)*, respectively. The bias for <sup>137</sup>Cs and <sup>241</sup>Am did not exceed 10-20%, for <sup>90</sup>Sr – 15-25%, for <sup>239+240</sup>Pu – 30%.

### RESULTS AND DISCUSSION

Ranges of values as well as the arithmetic mean and its error of activity concentration of <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>241</sup>Am, <sup>239+240</sup>Pu in components of the forest floor (organic and mineral constituent) and the top soil are listed in the table.

It was found that maxima of the activity concentration are characteristic of <sup>137</sup>Cs (up to 370 Bq/kg) and <sup>239+240</sup>Pu (up to 370 Bq/kg), lower concentrations are characteristic of <sup>90</sup>Sr (up to 190±30 Bq/kg), the content of <sup>241</sup>Am does not exceed 19±2 Bq/kg.

Based upon the in vitro analysis, it was found that activity concentrations of radionuclides are on average higher in the forest floor than in soil: the fate of the content of radionuclides was 71% in the floor (39% in the organic and 32% in the mineral constituent) and 29% in soil. The distribution of radionuclides among components of the forest floor and top soil is provided as histograms (Figure 2).

The minimum activity concentration in components of the forest floor and top soil is observed for <sup>241</sup>Am, the higher content was found for <sup>239+240</sup>Pu and <sup>90</sup>Sr, maxima are characteristic of <sup>137</sup>Cs. At the same time, the range of activity concentration values in the organic part of the floor for <sup>137</sup>Cs, <sup>90</sup>Sr and <sup>241</sup>Am is on average two orders of magnitude, for <sup>239+240</sup>Pu – three orders of magnitude; in the mineral constituent of the floor for <sup>90</sup>Sr and <sup>241</sup>Am is on average different by one order of magnitude, for <sup>137</sup>Cs and <sup>239+240</sup>Pu – two orders of magnitude; in the top soil, differences in ranges of activity concentration values average one order of magnitude for <sup>241</sup>Am and <sup>90</sup>Sr, two orders of magnitude for <sup>137</sup>Cs and three orders of magnitude for <sup>239+240</sup>Pu.

Based on data obtained, a series according to the accumulation degree of <sup>137</sup>Cs was arranged: the mineral constituent of the floor > top soil > organic part of the floor. The cause of the low vertical migration of <sup>137</sup>Cs is that it is tightly fixed in the soil absorbing complex. A lot of literature data on most of forest soils proves that the radionuclide is mostly accumulated in the lower layer of the forest floor or in top soil, which is proved by the data obtained in the course of research.

Table. Activity concentrations of artificial <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>241</sup>Am, <sup>239+240</sup>Pu in components of the forest floor and the top soil

Components under study	Activity concentrations of radionuclides, Bq/kg *)			
	<sup>137</sup> Cs	<sup>90</sup> Sr	<sup>241</sup> Am	<sup>239+240</sup> Pu
Organic part of the floor	$\frac{42 \pm 13}{1,1 - 370 (n - 33)}$	$\frac{30 \pm 7}{1,4 - 190 (n - 33)}$	$\frac{3 \pm 1}{0,3 - 19 (n - 21)}$	$\frac{34 \pm 16}{0,3 - 320 (n - 31)}$
Mineral part of the floor	$\frac{63 \pm 15}{2,7 - 350 (n - 32)}$	$\frac{12 \pm 2}{1,4 - 48 (n - 21)}$	$\frac{3 \pm 1}{0,9 - 9,7 (n - 19)}$	$\frac{66 \pm 28}{1,9 - 370 (n - 21)}$
Top soil	$\frac{36 \pm 5}{4,3 - 120 (n - 31)}$	$\frac{17 \pm 4}{1,1 - 71 (n - 25)}$	$\frac{2 \pm 0,3}{0,5 - 5,1 (n - 20)}$	$\frac{33 \pm 10}{0,7 - 240 (n - 28)}$

\*) In the numerator – arithmetic mean, error of the arithmetic mean; in the denominator – the minimum and maximum activity concentration; n – the number of test samples

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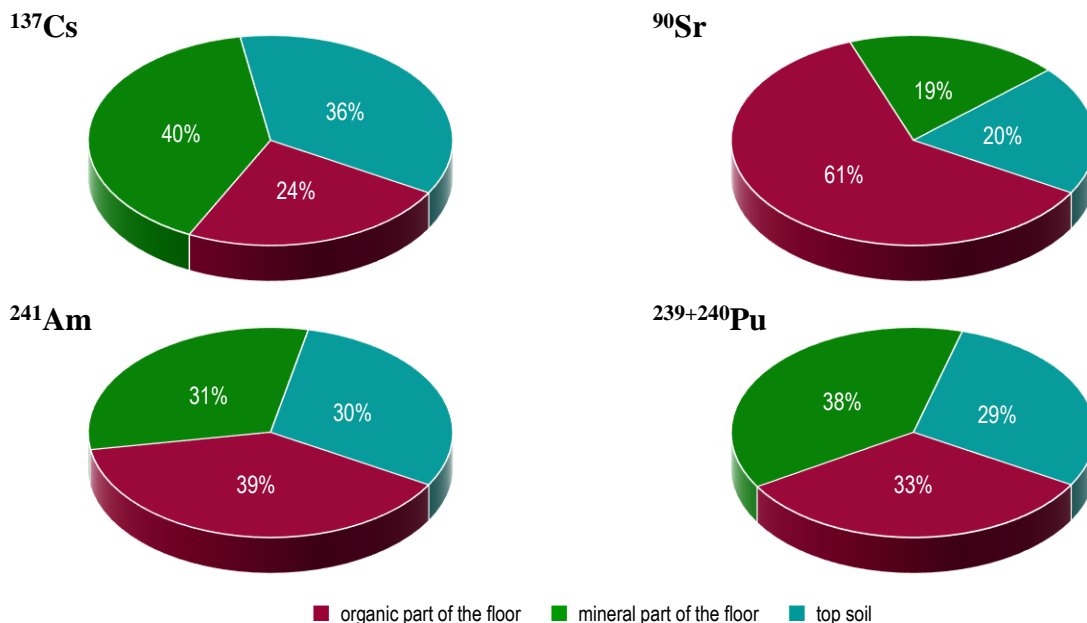


Figure 2. The average content of <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>241</sup>Am and <sup>239+240</sup>Pu in components of the forest floor and top soil

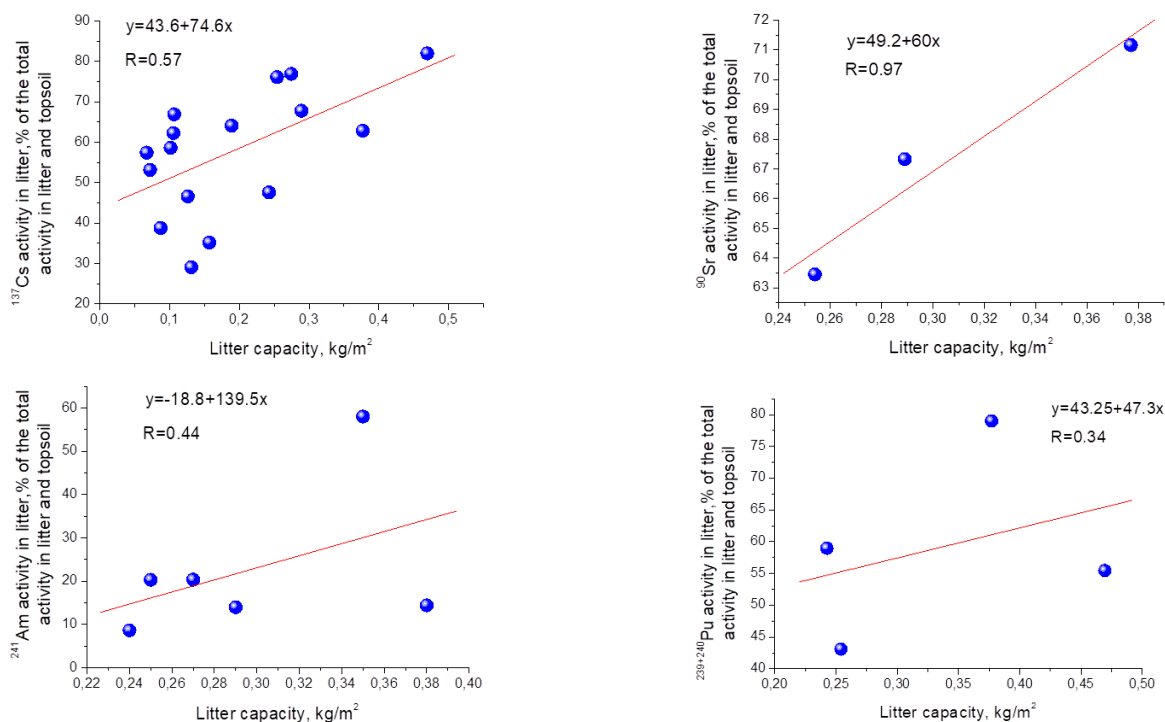


Figure 3. A variation in the inventory of radionuclides in the floor depending on its thickness

<sup>90</sup>Sr behaves different, the series of which according to the accumulation degree appears as follows: the organic part of the forest floor > top soil > mineral constituent of the forest. It is known that most of <sup>90</sup>Sr in natural components are oftentimes in mobile forms (water-soluble and exchangeable). A high index of easily accessible form of <sup>90</sup>Sr defines its intensive migration ability, which indicates its higher mobility and, consequently, a higher biological availability for plants. This can explain an intensive accumulation of <sup>90</sup>Sr in the

organic part of the floor.

For radionuclides of the transuranic series, the following series according to the accumulation degree were arranged: for <sup>241</sup>Am: the mineral constituent of the forest floor > the organic part of the floor > top soil; for <sup>239+240</sup>Pu: the organic part of the floor > the mineral constituent of the floor > top soil. As a result, research undertaken, it was found that the major inventory of transuranic elements (<sup>241</sup>Am, <sup>239+240</sup>Pu) is contained in the floor. At the same time, most of <sup>241</sup>Am, <sup>239+240</sup>Pu is

incorporated in slightly-soluble complex compounds, which explains their low mobility and accumulation in the organic and mineral constituent of the floor.

A special role of the forest floor in accumulation processes of radioactive substances is largely defined by its structure and thickness – the weight of the floor per unit area. The average thickness of the floor for the territory of interest is 0.25 kg/m<sup>2</sup>. A variation in the inventory of radionuclides in the floor depending on its thickness is listed in graphs (Figure 3).

As you see from the graphs provided, an increase of the accumulation of radionuclides in the forest floor is observed as its thickness increases, which, in turn, proved a special role of the floor in accumulation and distribution processes of radionuclides in the forest floor as a whole.

#### CONCLUSION

As a result of the researches conducted, concentrations of <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>241</sup>Am and <sup>239+240</sup>Pu in the top layer of soil as well as in mineral and in organic components of litter were determined. The maximum values of specific

activity were registered for <sup>137</sup>Cs (up to 370 Bq/kg) and <sup>239+240</sup>Pu (up to 370 Bq/kg), lower concentrations were registered for <sup>90</sup>Sr (up to 190±30 Bq/kg). Concentration of <sup>241</sup>Am did not exceed 19±2 Bq/kg.

Based on the study of radionuclide distribution in the top soil and components of the forest floor, series were arranged according to accumulation abilities of radionuclides: <sup>137</sup>Cs: the mineral constituent of the floor > top soil > the organic part of the floor; <sup>90</sup>Sr: the organic part of the floor > top soil > the mineral constituent of the floor; <sup>241</sup>Am: the mineral constituent of the floor > the organic part of the floor > top soil; <sup>239+240</sup>Pu: the organic part of the floor > the mineral constituent of the floor > top soil. It was also found that the activity concentration of radionuclides in on average higher in the forest floor than in soil: the fate of the content of radionuclides was 71% in the floor (39% in the organic and 32% in the mineral constituent) and 29% in soil. At the same time, an increase in the accumulation of radionuclides by the forest floor depends on its thickness.

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### СЕМЕЙ СЫНАҚ ПОЛИГОНЫНА ІРГЕЛЕС АУМАҚТАҒЫ ОРМАН ТӨСЕМІНДЕГІ ЖАСАНДЫ РАДИОНУКЛИДТЕР

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Мақалада Семей сынақ полигоны (ССП) аумағындағы ядролық сынақтардан кейінгі алыс кезеңдегі Ертіс маңындағы қарағайлы орманның беткі топырақ қабаты мен орман төсемінің құрауыштарындағы жасанды радионуклидтердің құрамы мен таралуын зерттеу нәтижелері ұсынылған. Зерттеліп жатқан топырақтарда, сондай-ақ төсемнің минералды және органикалық құрауыштарындағы <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>241</sup>Am, <sup>239+240</sup>Pu меншікті белсенділігінің минималды және максималды мәндері анықталды. Радионуклидтердің беткі топырақ қабатында және орман төсемінің құрауыштарында жинақталу қабілеті бойынша қатарлары қалыптастырылған. Орман төсемінің құрауыштарындағы <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>241</sup>Am, <sup>239+240</sup>Pu радионуклидтерінің құрамы мен оның қуаты арасында байланыс орнатылды.

**Түйінді сөздер:** радиоэкология, Семей сынақ полигоны (ССП), жасанды радионуклидтер (<sup>137</sup>Cs, <sup>90</sup>Sr, <sup>241</sup>Am, <sup>239+240</sup>Pu), орман экожүйесі, орман төсеміші.

**ИСКУССТВЕННЫЕ РАДИОНУКЛИДЫ В ЛЕСНОЙ ПОДСТИЛКЕ НА ТЕРРИТОРИИ,  
ПРИЛЕГАЮЩЕЙ К СЕМИПАЛАТИНСКОМУ ИСПЫТАТЕЛЬНОМУ ПОЛИГОНУ**

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В статье представлены результаты исследования содержания и распределения искусственных радионуклидов в компонентах лесной подстилки и верхнем почвенном слое соснового бора Прииртышья, в отдаленный период после ядерных испытаний на территории Семипалатинского испытательного полигона (СИП). Определены минимальные и максимальные значения удельной активности  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$ ,  $^{239+240}\text{Pu}$  в исследуемых почвах, а также в минеральной и органической составляющей подстилки. Сформированы ряды радионуклидов по их способности к накоплению в верхнем почвенном слое и компонентах лесной подстилки. Установлена зависимость между содержанием радионуклидов  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$ ,  $^{239+240}\text{Pu}$  в компонентах лесной подстилки и ее мощностью.

**Ключевые слова:** радиоэкология, Семипалатинский испытательный полигон (СИП), искусственные радионуклиды ( $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$ ,  $^{239+240}\text{Pu}$ ), лесная экосистема, лесная подстилка.