

# MEN-MADE RADIONUCLIDES IN LICHENS AND MOSSES OF THE KHARKIV REGION

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The specific activity of radionuclides in the soil, mosses, lichens, foliar trees and phytomass of a pine has been registered. The peculiarities of migration and accumulation of <sup>137</sup>Cs, <sup>40</sup>K, <sup>226</sup>Ra radionuclides in soil and plants were found. Investigations showed that the specific activity, for example, of <sup>137</sup>Cs ( $2 \cdot 10^{-2}$  Bq/g) in soil, ( $5.25 \cdot 10^{-3}$  Bq/g) in phytomass of a pine and ( $3.51 \cdot 10^{-2}$  Bq/g) in mosses and lichens was more, than in foliage trees ( $1.44 \cdot 10^{-3}$  Bq/g). These results can be used for estimation of radionuclides in the soil and plants in Kharkiv region.

PACS: 89.60.Gg

## INTRODUCTION

Currently, considerable the factual material about the intensity of the radioactive fallout and the content of individual radionuclides on the earth's surface was accumulated. These data are widely published, but available information not compiled and analyzed enough. In the Kharkiv region similar results are even less representative [1 - 5].

The difference between the amount of the fallout of fission products and the accumulation of them in the soil cover are not fixed and vary quite widely. This is due to the influence of temporary factors on radionuclides migration. Firstly, the fallout of <sup>90</sup>Sr, <sup>95</sup>Zr, <sup>106</sup>Ru, <sup>137</sup>Cs, <sup>144</sup>Ce are not only in the form of cations but also in the form anion and neutral forms. It is known that the products of fission in anionic form connect with the soil poorly and quickly migrate into the soil profile. Secondly, the physical and chemical properties of the soil play a role in the redistribution of radionuclides in soil cover [6].

The fallout of radionuclides on a surface of the soil due to ionic adsorption is trapped by anions of a crystal lattice of minerals. Significant impact on the behavior of radionuclides in the soil have cations  $\text{NH}_4^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{H}^+$ , which increase their mobility and anions  $\text{SO}_4^{2-}$ ,  $\text{PO}_4^{3-}$ , that reduce their mobility. In the latter case, the formation of poorly soluble compounds takes place for such as <sup>90</sup>Sr and <sup>137</sup>Cs.

Analogous elements for <sup>90</sup>Sr-Ca and for <sup>137</sup>Cs-K are influenced on the kinetics of sorption of radionuclides also.

The soil, mosses, lichens, foliar trees and phytomass of a pine were researched as objects of accumulation of radionuclides. The variety in the choice of objects of study is caused by the fact that these objects are characterized by different capacity for removal and accumulation of radionuclides from soil, as well as selectivity in relation to them.

The indicator properties of mosses and lichens well known because of them accumulate different radionuclides. The aboveground part of the woody plants can take up to 90% of the falling radionuclides, which can then migrate to the root system. Foliar trees detain fewer of radionuclides than phytomass of a pine, which effectively accumulate radionuclides throughout the year [7, 8].

The aim of this work was to study soil contamination by radionuclides and accumulation of radionuclides in some plant objects in Kharkiv region.

## 1. MATERIALS AND METHODS

The studies were conducted with soil samples and plant objects (mosses, lichens, foliar trees, phytomass of pine), which were collected in the Kharkov region. Preparations samples for analysis included:

1. Ashing of soil at a temperature 450°C during 12 hours;
2. Drying and grinding plant samples;
3. Ashing of plant facilities in quartz bowls in an oven, first at a temperature 105°C (charring) and then at 450...520°C (samples to complete mineralization);
4. Preparations of standards.

Specimens (weight of at least 1 g) and standards were placed in aluminum foil for  $\gamma$ -activating by bremsstrahlung from linac with  $E=12.5$  MeV and a current of 700  $\mu\text{A}$ . Measurement of the elements after  $\gamma$ -activating, as well as the activity of transuranic radioactive fallout was conducted by a Ge(Li)-detectors with energy resolution of 3.2 at 1333 keV.

## 2. RESULTS AND DISCUSSION

Time of measurements of soil samples was from 24 to 28 hours. The specific activity of <sup>137</sup>Cs in the upper layer of soil was  $2 \cdot 10^{-2}$  Bq/g. <sup>137</sup>Cs soil contamination density due to global radioactive contamination in 1986 was  $3.0 \dots 4.7$  kBq/m<sup>2</sup> with an average value  $3.9 \pm 0.8$  kBq/m<sup>2</sup> [9].

The measurement results of the <sup>40</sup>K specific activity in the soil, which is a chemical analog of <sup>137</sup>Cs, showed values in the level  $\sim 6 \cdot 10^{-1}$  Bq/g. The exchange for Cs<sup>+</sup> and K<sup>+</sup> ions in the soil is a dynamic reaction. The direction of the reaction can easily be displaced by little change in the concentration of these elements. The bond strength atoms of soil colloids of K lower than for Cs atoms in this case. Therefore, the exchange sorption for Cs<sup>+</sup> and K<sup>+</sup> in the soil is preferably towards for formation of compounds Cs<sup>+</sup>-soil. <sup>137</sup>Cs sorption kinetics depend on the soil properties, the degree of hydration, as well as some differences in the ionic radii Cs (0.169 nm), K (0.092 nm). For example, the soil which saturated with K<sup>+</sup> and Ca<sup>2+</sup> absorb of <sup>137</sup>Cs most strong-

ly, and a state of equilibrium is reached fairly quickly. Uptake of these ions in plant objects is largely dependent on the degree of hydration of the soil. The addition of  $K^+$  ions in the soil decreases uptake of  $^{137}Cs$  into plants. The increase  $H^+$  ions soil contributes to the appearance of significant amounts of mobile forms  $^{137}Cs$ , which are at low pH~4, are transported into the plant [10].

In our studies, the specific activity of  $^{226}Ra$  was measured, which amounted to  $6 \cdot 10^{-2}$  Bq/g.  $^{226}Ra$  source in the atmosphere are the soil particles in the air in a suspended state. It is known that each gram of  $^{137}Cs$  activity has ~90 Ci, or decay rate equal to the decay rate of ~90 g radium.

$^{226}Ra$  found in drinking water and ~10% of this radionuclide uptake in the human body of water.

Error in measuring the specific activity of the radionuclides in the soil was ~15%.

The extremely important role of radioecological monitoring of region is radionuclide content in lichens. Lichens do not have the root system, but they effectively adsorb of radionuclides from the upper layer of most contaminated soil. The such long-life radionuclides as  $^{55}Fe$ ,  $^{144}Ce$ ,  $^{65}Zn$ ,  $^7Be$ , isotopes of U et al. accumulate by lichens in addition to  $^{90}Sr$ ,  $^{137}Cs$ ,  $^{210}Pb$ ,  $^{210}Po$  [11].

Fig. 1 shows the  $\gamma$ -spectrum of mosses and lichens.

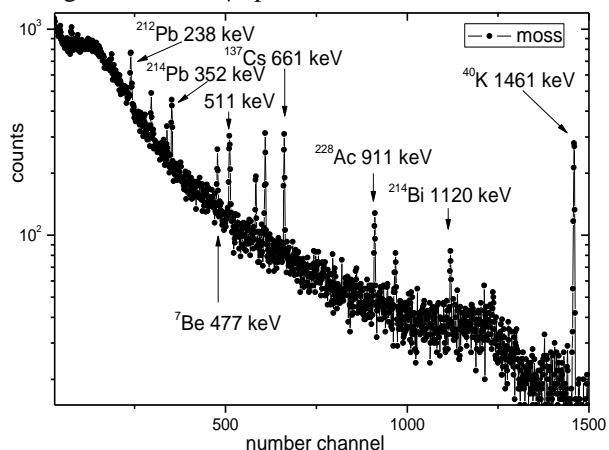


Fig. 1. The spectrum of mosses and lichens after irradiated bremsstrahlung with  $E_{max} = 12.5$  MeV

The values of the specific activity of certain radionuclides contained in mosses and lichens are presented in Table 1.

Table 1

The specific activity in mosses and lichens (mBq/g)

Object	$^{137}Cs$	$^{40}K$	$^7Be$	$^{226}Ra$
Mosses-lichens	35.1	422.0	130.0	15.0

The error of measurement of the specific activity of radionuclides in mosses and lichens is ~15%.

The specific activity of  $^{137}Cs$  in mosses and lichens is ~1.7 times higher than in the soil. Despite the ability of lichen to convert sparingly soluble radionuclides in easily movable form and to increase their migration ability, they also can accumulate and hold them in large amounts. The main accumulation of  $^{137}Cs$  and  $^{90}Sr$  in mosses and lichens comes from atmospheric deposition [12].

The content of  $^{40}K$  in mosses and lichens is in ~1.5 times higher than in the soil. As a result, of the highly-developed surface, the moss-lichen system is an effective sorbent, and a low level of metabolic processes promotes the accumulation of biomass in the radionuclides. Potassium is an essential trace element in the metabolic processes and vital activity of mosses and lichens. The system of mosses-lichens absorbs radionuclides of airborne soil particles and the absorption of liquid atmospheric precipitation (rain, snow, etc.). In soil and air, the specific activity of  $^{40}K$  is also superior to the specific activity of  $^{137}Cs$ ,  $^{226}Ra$ , and  $^{232}Th$ .

Mosses and lichens contain  $^7Be$  often.  $^7Be$  is cosmogenic radionuclides and often found in air and precipitation [13].

Exceeding of the specific activity of  $^{226}Ra$  in soil in ~4 times relatively of the radionuclide content in mosses and lichens can be associated with the proximity of the situation of TPP, coal-fired. It is well known that the global average specific activity of  $^{226}Ra$  in the fly ash TPP is ~240 Bq/kg.

The values of the specific activity of radionuclides in foliar trees and phytomass of a pine are presented in Table 2.

Table 2

The specific activity of radionuclides in foliar trees and phytomass of a pine (mBq/g)

Object	$^{137}Cs$	$^{40}K$	$^{228}Ac$	$^{226}Ra$	$^{210}Pb$
Foliar trees	1.8	238	0.90	1.9	1.02
Phytomass of a pine	5.5	40.2	3.9	7.0	3.9

The error of measurement of the specific activity in the radionuclides of foliar trees and phytomass of a pine is ~20%.

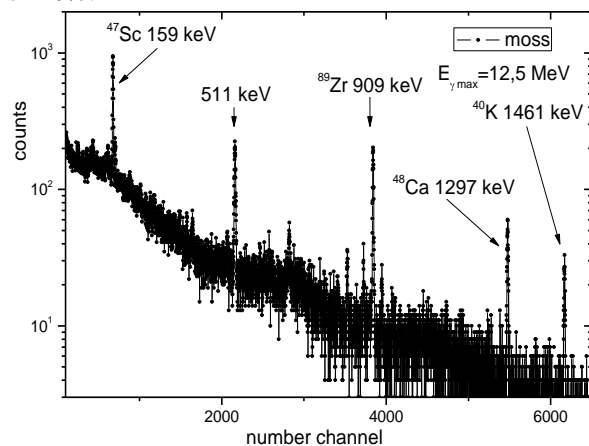


Fig. 2. The spectrum of moss and lichens after irradiated bremsstrahlung with  $E_{max} = 12.5$  MeV

The results show a higher bioavailability of  $^{137}Cs$ ,  $^{226}Ra$ ,  $^{228}Ac$  and  $^{210}Pb$  for phytomass of a pine compared with accumulation those radionuclides into foliar trees.

Higher ability of the accumulation of radioactive aerosols by phytomass of a pine as compared with foliar trees is connected with the high radiosensitivity of softwood [14]. Also the evergreen needles of a pine accumulate radionuclides during year.

The Fig. 2 shows the  $\gamma$ -spectrum of moss has been irradiated by bremsstrahlung with maximum energy of 12.5 MeV.

Table 3  
The ratio of the element content *R* in different objects (dry matter)

Element	R, moss /soil	R, leaf celandine/soil	R, root celandine/soil	R, moss/celandine
Ca	1.7	1.53	1.8	1.06
Ti	1.07	0.02	0.14	14.02
Mn	1.17	0.06	0.35	5.66
As	0.92	0.04	0.12	11.36
Br	1.28	–	–	–
Rb	0.76	0.16	0.22	4.03
Zr	0.65	0.01	0.13	9.45
Nb	0.98	0.05	0.35	4.92
I	1.55	0.48	0.58	2.92
Cs	0.31	0.04	0.15	3.24
Ce	0.58	0.01	0.11	9.75
Pb	2.63	0.33	0.32	8.09
U	0.80	0.14	0.10	6.59

The measurement error regarding the content of elements in moss and celandine is ~15%

Table 4  
The ratio of the radionuclide content *R* in different objects (dry matter)

Isotope	R, moss/soil	R, celandine/soil	R, moss/celandine
<sup>40</sup> K	1.25	4.79	0.26
<sup>137</sup> Cs	11.83	0.10	121.46
<sup>232</sup> Th	1.43	0.24	5.96
<sup>228</sup> Ac	1.85	0.29	–
<sup>212</sup> Pb	1.18	0.18	–
<sup>208</sup> Tl	1.26	0.25	–
<sup>238</sup> U	1.27	0.46	2.79
<sup>226</sup> Ra	0.64	0.06	–
<sup>214</sup> Pb	1.25	0.29	–
<sup>214</sup> Bi	1.92	1.02	–

Error ratio measurement of the specific activity of radioactive elements in moss and celandine is ~25%.

It has been shown (Tabl. 3), that the transfer factor soil-to-plant for celandine is in the conventional range of most common elements [15]. The transfer factor soil-to-plant of celandine for calcium, iodine and lead necessary is rather high. The transfer factor soil-to-plant of moss has quite a high value. [16] The highest transfer factor soil-to-plant observed for <sup>210</sup>Po and <sup>210</sup>Pb in moss, which make up 25...50 [16]. Also, there are relatively high transfer factor soil-to-plant in moss compared with celandine for elements which do not participate in the metabolism of living organisms: Ti, As, Zr, Ge, Pb, U. Various content of potassium in moss and celandine lead to a huge difference in transfer factor soil-to-plant for <sup>137</sup>Cs (Tabl. 4).

## CONCLUSIONS

1. The content of elements in the soil, mosses, lichens, foliar trees and phytomass of a pine after activated, as well activity of transuranic and radioactive elements were measured.

2. The differences and peculiarities in the accumulation of <sup>137</sup>Cs, <sup>40</sup>K, <sup>226</sup>Ra and other isotopes in researched objects, taken in the Kharkov region, were discovered.

3. It is shown that the values of the specific activity, such as <sup>137</sup>Cs in the soil ( $2 \cdot 10^{-2}$  Bq/g), phytomass of a pine ( $5.25 \cdot 10^{-3}$  Bq/g), and lichens, mosses ( $3.51 \cdot 10^{-2}$  Bq/g) was higher than in foliar trees ( $1.44 \cdot 10^{-3}$  Bq/g), which can be explained by the different radiosensitivity of these objects, as well as the different mechanisms of uptake and accumulation of these radionuclides.

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Article received 14.03.2016

#### **ТЕХНОГЕННЫЕ РАДИОНУКЛИДЫ В ЛИШАЙНИКАХ И МХАХ ХАРЬКОВСКОГО РЕГИОНА**

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Измерена удельная активность радионуклидов в почве, мхах, лишайниках, лиственных деревьях и хвойной фитомассе. Изучены особенности поступления и накопления радионуклидов  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  и др. в исследуемые объекты. Показано, что значения удельной активности, например для  $^{137}\text{Cs}$ , в почве ( $2 \cdot 10^{-2}$  Бк/г), хвойной фитомассе ( $5,25 \cdot 10^{-3}$  Бк/г) и лишайниках-мхах ( $3,51 \cdot 10^{-2}$  Бк/г) были выше, чем в лиственных деревьях ( $1,44 \cdot 10^{-3}$  Бк/г). Подобная тенденция сохраняется и для других радионуклидов. Результаты могут быть использованы для оценки содержания радионуклидов в почве и различных растительных объектах Харьковского региона.

#### **ТЕХНОГЕННІ РАДІОНУКЛІДИ В ЛИШАЙНИКАХ І МОХАХ ХАРКІВСЬКОГО РЕГІОНУ**

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Проведено вимірювання питомої активності радіонуклідів у ґрунті, моховинні, лишайниках, листяних деревах та хвойній фітомасі. Вивчені особливості надходження і накопичення радіонуклідів  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  та інших у досліджувані зразки. Показано, що значення питомої активності, наприклад для  $^{137}\text{Cs}$ , у ґрунті ( $2 \cdot 10^{-2}$  Бк/г), хвойній фітомасі ( $5,25 \cdot 10^{-3}$  Бк/г) та моховинні і лишайниках ( $3,51 \cdot 10^{-2}$  Бк/г) були вищими, ніж у листяних деревах ( $1,44 \cdot 10^{-3}$  Бк/г). Подібна тенденція зберігалась і для інших радіонуклідів. Результати можуть бути використані для оцінки складу радіонуклідів у ґрунті та інших рослинних об'єктах Харківського регіону.