UDC (504.054:546.79):630.1:582.912.46:477.42 DOI: https://doi.org/10.33730/2077-4893.1.2024.299940

DYNAMICS OF RADIOACTIVE CONTAMINATION OF LINGONBERRY (*VACCINIUM VITIS-IDAEA* L.) IN THE FORESTS OF ZHYTOMYR POLISSIA (UKRAINE) SINCE THE CHORNOBYL NUCLEAR ACCIDENT

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У статті наведені та проаналізовані матеріали вивчення рівнів радіоактивного забруднення ґрунту та питомої активності ^{137}Cs у надземній частині фітомаси та ягодах брусниці на постійних пробних площах у різні роки з часу аварії на Чорнобильській атомній станції. В дослідженнях, які виконані у Поліському філіалі Українського науково-дослідного інституту лісового господарства та агролісомеліорації впродовж 1998–2021 рр., використовувались загальнонаукові та математико-статистичні методи, а також порівняльний аналіз. З метою обґрунтування досліджень використані звітні дані лісогосподарських підприємств Волинської, Рівненської та Житомирської обл. Показано, що відбувається істотне зменшення щільності радіоактивного забруднення ґрунту, а також питомої активності ^{137}Cs у надземній частині фітомаси та ягодах брусниці у лісах регіону дослідження за період спостережень. Виявлено, що нині основна частка сумарної активності радіонукліду знаходиться у верхньому мінеральному 10 см шарі ґрунту — 74,7%. Встановлено, що у надземній частині фітомаси брусниці зберігається значний вміст радіонукліду навіть при невеликій щільності радіоактивного забруднення ґрунту. З'ясовано, що у 1998 р. за мінімальної величини щільності забруднення ґрунту ^{137}Cs (49 ± 5,6 кБк·м$^{-2}$) питома активність ^{137}Cs у надземній фітомасі брусниці становила 8043 ± 510,9 Бк·кг$^{-1}$, тоді як у 2021 р. (мінімальній величині щільності забруднення ґрунту ^{137}Cs 22 ± 0,3 кБк·м$^{-2}$) — 1046 ± 182 Бк·кг$^{-1}$. Виявлені лінійні залежності між щільністю радіоактивним забрудненням ґрунту та вмістом ^{137}Cs в ягодах і надземній частині фітомаси брусниці, які можна використовувати у практиці їх заготівлі.

Ключові слова: *щільність забруднення ґрунту ^{137}Cs, питома активність, радіонуклід, ягідні рослини, радіаційна токсикологія, лісові екосистеми.*

INTRODUCTION

As a result of the Chornobyl Nuclear Power Plant accident forests experienced significantly high level of radioactive contamination compared to open landscapes. The largest areas and levels of this contamination were observed in the wood areas of Polissia region in our country. This can be explained by the processes which occurred in the destroyed reactor and due to the weather conditions as well in the period of the most intensive release of radionuclides in the environment. In the most forested part of Polissia, including Volyn, Rivne, and Zhytomyr regions the area of forests with a soil contamination density of ^{137}Cs exceeding 37 kBq·m^{-2} (1 Ci·km^{-2})

in 1991 was 860 000 hectares. Thus, forests accumulated a remarkable amount of radionuclides and they became a source of radiation hazard for the local population [1].

At the same time the forested areas of the marked regions are rich for berry plants, which have been long used by the local population for personal consumption and for sale. In the beginning of the first months since the Chornobyl Nuclear Power Plant accident, the harvesting of wild berries in Ukrainian forests were prohibited [2]. Over time, after surveying forests for radioactive contamination and obtaining the results of initial studies on the content of radionuclides in wild berry plants, regulated in accordance with the established limitation at that time [3]. Further this regulation was actualized in connection with the receipt of new materials on the radiation situation in forests and the migration of radionuclides to different species of berry plants based on forest-typological approach.

Over the past 20 years, studies on the migration of radioactive elements in forest ecosystems and their entry into various components, including berry plants, have highly decreased and had a fragmentary character. Meanwhile, with the purpose of conducting radiation toxicology dose calculations, define potential areas for wild berry harvesting, forecasting levels of their radioactive contamination and rehabilitating forest plantations, data on the accumulation of ^{137}Cs in different types of forest ecological conditions are necessary. Hereby remarked limitations, these studies remain relevant.

The purpose of the research was determination of the current levels of radioactive contamination of the common forest berry plant — lingonberry (*Vaccinium vitis-idaea* L.), which are wide-spread in Ukrainian forests of Polissia, in different types of forest ecological conditions and in stands of different tree species composition.

ANALYSIS OF RECENT RESEARCH

Fragmentary studies on the radioactive contamination of wild berry plants, including lingonberry were conducted in Ukraine and some other European countries in the 1960s–1980s. In the Ukrainian Carpathian Mountains researchers investigated the ^{137}Cs content in the organs of some berry dwarf-shrubs, such as lingonberry, bilberry (*Vaccinium myrtillus* L.) and blueberry (*Vaccinium uliginosum* L.) [4]. They found that ^{137}Cs activity concentration in lingonberry organs was higher than in bilberry and blueberry. In all studied species this indicator was the highest in the plant shoots and the lowest — in the berries. Somewhat different results were obtained later by German researchers who ranked the organs of berry plants in terms of their ^{137}Cs content as follows: flowers > leaves > berries > roots > shoots [5]. Similar results were obtained in 2015 by researchers in Volyn Polissia where lingonberry organs, according to ^{137}Cs content, were ranked as follows: berries > leaves > roots > shoots [6].

In post-Chornobyl period a remarkable number of publications in scientific literature emerged, dedicated to study peculiarities of ^{137}Cs accumulation in various plants of forest ecosystems, including berries. This was prompted by the wide use of these plants' berries for consumption and the production of medicinal preparations in many European countries. Researchers in several European countries classified lingonberry as a plant with significant accumulation of ^{137}Cs in berries [7–11]. Lingonberry, unlike bilberry, belongs to plants in which the absorption capacity of ^{137}Cs does not change depending on the levels' industrial pollution of the environment [12], and later observations by other researchers showed opposite results [13]. With increasing of anthropogenic pollution, the ^{137}Cs content in lingonberry decreased. The reduction of ^{137}Cs in lingonberry was also supported by active experiments on the application of various amounts of wood ash [14].

Extensive and multi-faceted radioecological studies of lingonberry were conducted in Finland [15]. Scientists found that lingonberry has significantly higher ^{137}Cs content in various parts and organs compared to other species growing in the same ecological conditions. They observed an increase in the inten-

sity of radionuclide influx in this berry plant over three years since radioactive fallouts on Finnish forest ecosystems. Researchers concluded that lingonberry berries could play a significant role in the accumulation of ^{137}Cs in the human body, and the consumption of wild berries in substantial quantities could have a certain toxic effect on the human body. Somewhat different results were obtained by Swedish scientists who studied the ^{137}Cs content in various plant species over a much longer period — from 1986 to 1994 [16]. They concluded that all plants, including lingonberry, showed the gradual decrease in ^{137}Cs activity concentration in the aboveground parts of the plants.

Comparing the indicators which characterize the intensity of ^{137}Cs influx into berries and the vegetative mass of lingonberry shown by different authors demonstrates significant differences. This can be explained by various factors such as different years of research, variations in the ecological conditions of plant growth, the tree species composition of the forest stands in the study area, methodological differences, etc. However, studies conducted in European forests allowed researchers to generalize that the part of forest food products (edible mushrooms, wild berries) in internal radiation exposure dose on the local population can reach 80% of the total dose obtained from all food products [17; 18].

Broader research on study the radioactive contamination of wild berry plants in general, and lingonberry in particular, was conducted in Ukraine. Researchers studying interspecies peculiarities in the accumulation of ^{137}Cs by berry plants classified lingonberry as an intensive radionuclide accumulator [19]. Studies conducted in various types of forest ecological conditions allowed researchers to conclude that the intensity of ^{137}Cs influx into different parts and organs of lingonberry was the lowest in fairly fertile site type and the highest — in infertile pine site type [20]. Scientists concluded that this is explained by the non-exchangeable fixation of the radionuclide in the soil in more favorable growth conditions. Researchers also identified the accumulation of ^{137}Cs by lingonberry dwarf-shrubs of different ages [21] and the peculiarities of radionuclide influx during the vegetative period [22]. It was found that the maximum values of the activity concentration of ^{137}Cs in lingonberry phytomass was observed in May–June, in the further periods of the growing season, its gradual decrease is observed. Ukrainian scientists also investigated the changing in the levels' radioactive contamination of lingonberry berries during short observation periods and noted some reduction in the levels' radioactive contamination of lingonberries in the 1990s [17]. Results from more extended studies on the levels of radioactive contamination of lingonberry are currently unavailable.

MATERIALS AND METHODS OF RESEARCH

Lingonberry is widespread in coniferous, mixed and deciduous forests, as well as in open areas (clearings, forest glades) in types of forest conditions, ranging from fresh to wet infertile pine, fairly infertile pine and fairly fertile site type. Depending on the of forest site type, rhizomes of lingonberry are located between the mineral part of the soil and the forest litter, or in the upper part of the peat. The depth of penetration of additional roots into the soil in autotrophic conditions is 3–10 cm, and under hydromorphic ones, can reach up to 25 cm

Research was conducted on permanent experimental plots on the territory of the branch «Luhynske Forest Enterprise» of the State Enterprise «Forests of Ukraine» (Zhytomyr region) in pine plantations with sufficiently close forestry and ecological characteristics in moist fairly infertile pine site type (B_3). Now they presented by pure pine stands aged 65–75 years with a relative completeness of 0.8–0.9, a stand quality class — I. Undergrowth consisted of individual specimens of *Pinus sylvestris* L. and *Betula pendula* Roth. The understory consisted of *Sorbus aucuparia* L. and *Frangula alnus* Mill. The herbaceous-dwarf-shrub layer was dense and consisted on *Vaccinium myrtillus* with a projected coverage of 50–60%, *V. vitis-idaea* —

10–15%, *Pteridium aquilinum* (L.) Kuhn. — 1–5%, *Molinia caerulea* (L.) Moench — 1–3% and *Lycopodium clavatum* L. Moss layer was well-developed and consisted on *Pleurozium schreberi* (Wild. ex Brid.) Mitt. — 45–55% and *Dicranum polysetum* Sw.) — 40–50%. The soil is soddy-middle podzol, sandy-loam. The association on all experimental plots was Pineturn myrtilloso-hylocomiosum.

On each experimental plot during the berry ripening period 5 accounting plots with an area of 1 m^2 (1 × 1 m) were established, from which soil samples (using the coring method) were collected to determine the density of ^{137}Cs soil contamination (A_s). Additionally, samples of berries and aboveground biomass of lingonberry were collected to measure the activity concentration of ^{137}Cs (A_m). In laboratory conditions samples of soil and aboveground biomass were dried to an air-dry state at temperature of 80°C during 72 hours, then ground and homogenized. Berries were analyzed in fresh state. The activity concentration of ^{137}Cs was measured using the LP-4900B «AFORA» spectrum analyzer with GeLi-detector DGDK-100B3 and also spectrum analyzers SEG-001 «AKP-C»-63 and SEG-001 «AKP-C»-150. The measurement error of ^{137}Cs activity concentration was less than 15% (confidence level — 0.95). The statistical processing of research results was carried out using standard software packages such as Database DB2, «Statgrafics», «Statistica» and «Microsoft Office Excel».

RESULTS AND DISCUSSION

Forest enterprises in the research region, which were subordinated to the Ministry of Forestry of Ukraine, before Chernobyl disaster, non-wood forest resources were extensively harvested, including wild berry plants, for their own processing as well as for selling to other enterprises (*Table 1*).

Forest enterprises gained significant profits from this activity. The radioactive contamination of the forests in 1986 led to the necessity of prohibiting or tightly regulating a certain part of the forestry activities of the region's enterprises, and the harvesting of wild berries fell under a complete ban.

The initial research on the scientific justification of using lingonberry in forests contaminated with radionuclides was conducted by us in 1998 with the aim of establishing dependencies between the density of radioactive soil contamination of ^{137}Cs and the activity concentration of the radionuclide in the berries and aboveground vegetative phytomass of lingonberry (widely used for food, and the latter — in official and folk medicine). It was found that ^{137}Cs enters its aboveground phytomass quite intensively, and even at small values of the first indicator — 49 ± 5.6 kBq·m^{-2}, ^{137}Cs activity concentration was high — 8043 ± 511 Bq·kg^{-1}.

The values of radionuclide transfer coefficients were quite noticeable and varied within pretty wide ranges: in the air-dry aboveground phytomass, from 33.4 to 164.1 $m^2 \cdot kg^{-1} \cdot 10^{-3}$, and in the fresh berries — from 9.5 to 26.0 $m^2 \cdot kg^{-1} \cdot 10^{-3}$. The obtained data allowed to categorize lingonberry as species that actively accumulates radionuclides. This may be explained by some biological characteristics of the species and, to some extent, by the ecological conditions of growth (weak non-exchangeable fixation of ^{137}Cs in sandy-loam soils of fairly infertile pine site type).

It was established that with the increase in radioactive soil contamination, the activity concentration of the radionuclide increases both in lingonberry berries and in the aboveground phytomass (*Fig. 1*). The content of ^{137}Cs in lingonberry berries and the aboveground phytomass depending on the density of radioactive soil contamination on permanent

Table 1. **Dynamics of harvesting volumes of wild berries by forestry enterprises in the forests of Volyn' and Zhytomyr Polissia**

Region	Harvest volumes by years, tons			
	1973	1978	1983	1987
Volyn'	244	997	1934	1689
Zhytomyr	229	157	602	1059
Rivne	1289	1456	4345	2734
Total	1762	2610	6881	5482

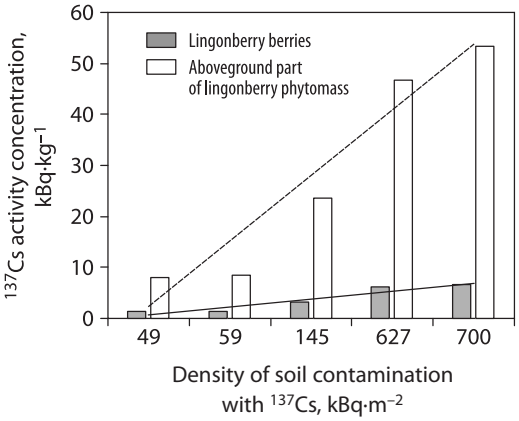

Fig. 1. Dynamics of ^{137}Cs activity concentration in fresh lingonberry berries and air-dry aboveground phytomass depending on the density of radioactive soil contamination on permanent experimental plots in 1998

experimental plots in 1998 was described by the following equations: for the air-dry aboveground phytomass of lingonberries: $A_m = 12.902 \cdot A_s - 10.658$ ($R^2 = 0.93$); for fresh berries: $A_m = 1.555 \cdot As - 0.907$ ($R^2 = 0.92$).

Over time, there was a redistribution of ^{137}Cs in forest ecosystems among their components: migration to greater depths in the soil with some fixation; influx into perennial plants, primarily tree species and concentration in them; a constant flow within a small biological cycle, ensuring the inflow and outflow of radionuclides to various components (including lingonberry). Additionally, there was a physical decay of the radioactive element both in the soil and in the biomass.

The materials obtained from the experimental plots in 2021 indicate that ^{137}Cs gradually migrates to the lower soil layers (*Table 2*). It was found that the major part of the total radionuclide activity is found in the mineral part of the soil — 86.3%, with 74.7% in the upper (10 cm) layers. It should be noted that the root system of lingonberry is located in this soil layer. It was shown that a part of the total activity of ^{137}Cs in the forest litter is quite substantial — 13.7%, which in turn, creates conditions for the accumulation of a certain amount of radionuclide in the upper,

Table 2. **The distribution of total activity of ^{137}Cs in the soil layers of moist fairly infertile pine site type (B_3) in 2021**

The soil layers	Total activity of ^{137}Cs	
	Bk·500 cm^{-2}	%
Forest litter		
O_l (undecomposed)	15	0.15
O_f (semi-decomposed)	554	5.53
O_h (decomposed)	801	8.00
Mineral soil layers		
0–2 cm	3281	32.76
2–4 cm	2182	21.79
4–6 cm	984	9.83
6–8 cm	656	6.55
8–10 cm	378	3.77
10–12 cm	286	2.85
12–14 cm	215	2.14
14–16 cm	137	1.37
16–18 cm	95	0.95
18–20 cm	90	0.90
20–22 cm	80	0.80
22–24 cm	61	0.61
24–26 cm	46	0.46
26–28 cm	35	0.35
28–30 cm	39	0.39
30–32 cm	20	0.20
32–34 cm	19	0.19
34–36 cm	20	0.20
36–38 cm	12	0.12
38–40 cm	9	0.09
Total	**10016**	**100.0**

root-populated layer of the mineral part of the soil in order for the intensive influx to lingonberry.

The significant migration capacity of ^{137}Cs in the soddy-podzolic sandy-loam soils of the Polissia forests, and the possibility of its intensive entry into lingonberry are confirmed by the results of the study of radioactive contamination of lingonberry on experimental plots in 2021 (*Fig. 2*).

For instance, the activity concentration of the radionuclide in the aboveground phytomass on the experimental plot with the minimum density of radioactive soil contamina-

tion — 22.2 ± 0.25 kBq·m^{-2} reach in air-dry phytomass of 1046 ± 182 Bq·kg^{-1}, and in the fresh berries — 301 ± 27 Bq·kg^{-1}. The ^{137}Cs content in lingonberry berries indicates that at the low density of radioactive soil contamination it can be used as food, as it does not exceed the values of the «Permissible levels of radionuclide content of ^{137}Cs and ^{90}Sr in food and drinking water» — 500 Bq·kg^{-1} [23].

At the same time, the value of this indicator in the aboveground part of the designated sample area significantly exceeds the «Hygienic norm of ^{137}Cs and ^{90}Sr activity concentration in medicinal plant raw materials (substances) used for the production of medicinal products» — 500 Bq·kg^{-1} [24]. This allows us to conclude that at the present time, even with low values of the density of radioactive contamination of the soil there is an intensive accumulation of ^{137}Cs in the lingonberry phytomass in the moist fairly infertile pine site type (B_3).

The content of ^{137}Cs in the lingonberry depending on the density of radioactive soil contamination on permanent experimental plots in 2021 was described by the following equations: for the air-dry aboveground part of lingonberry phytomass: $A_m = 2.205 \cdot A_s - 2.515$ ($R^2 = 0.80$), and for fresh lingonberry berries: $A_m = 0.588 \cdot As - 0.628$ ($R^2 = 0.85$).

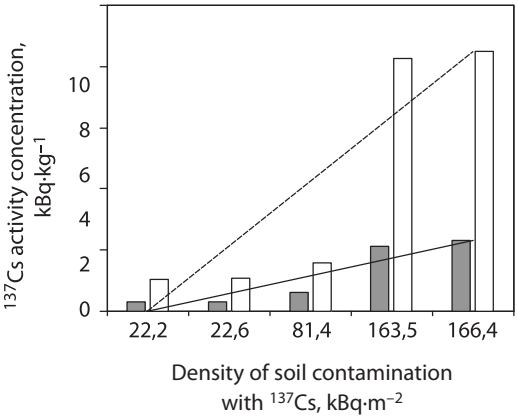

Fig. 2. Dynamics of ^{137}Cs activity concentration in lingonberry fresh berries and air-dry aboveground phytomass depending on the density of ^{137}Cs soil contamination on permanent experimental plots in 2021

CONCLUSIONS

During the studied period (1998–2021), a decrease in the density of radioactive soil contamination (2.4–3.1 times) is observed in the forest plantations of Polissia region of Ukraine. This can be explained by the decay of radionuclide and its migration into the components of forest ecosystems. The major part of ^{137}Cs total activity is found in the upper (10 cm) layer of the soil's mineral part — 74.7%, where the root systems of herbaceous and dwarf-shrub plants are concentrated in forest ecosystems.

It was found that lingonberry belongs to the group of plants characterized by a high content of ^{137}Cs in the aboveground vegetative phytomass. Harvesting it for the production of medicinal products should be restricted in all areas contaminated with radionuclides. Lingonberry fresh berries can be harvested in areas with a radioactive soil contamination density up to 74 kBq·m^{-2}.

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Стаття надійшла до редакції журналу 15.02.2024