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Radiological analysis of food products of forest origin in the pollution zone of the Kamin-Kashyrskyi district of the Volyn region of Ukraine

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Abstract: The problem of the radiological contamination of food products from secondary forest use remains relevant and requires targeted preventive measures, with the contamination being the result of the Chornobyl nuclear power plant disaster. The research was conducted with the aim of comprehensively monitoring the content of radionuclide ^{137}Cs in forest products (Wild Bilberry (*Vaccinium myrtillus*), Blackberries (*Rubus* L.), Wild Strawberries (*Fragaria vesca* L.), Raspberry (*Rubus idaeus* L.), together with *Cantharellus cibarius*, *Boletus edulis*, and *Leccinum scabrum* mushrooms) of the zone of radioactive contamination of the Kamin-Kashyrskyi district of the Volyn region. Samples were collected in 2020–2022. The total number of samples was 729 dried and 975 fresh mushrooms and berries samples for 2020, 1,154 dried and 886 fresh samples for 2021, and 870 dried and 896 fresh samples for 2022. The results of research indicate that in the specified territory, and at the present moment, forest products are to be found with a significant content of the radionuclide ^{137}Cs, including those exceeding the permissible levels. There is, therefore, a threat of further internal exposure of the local population via access to procurement points of contaminated products and restaurants. In connection with the detection of these pollutants in forest products in the studied area, further expansion of specialized radioecological monitoring studies and strengthening of radiological control remains relevant.

Keywords: radiological pollution, ^{137}Cs, ^{90}Sr, mushrooms, wild berries

INTRODUCTION

In the 37 years since the accident at the Chornobyl Nuclear Power Plant, the volumes of monitoring of the migration of radioactive elements in forest ecosystems and the levels of pollution of forest products have significantly decreased, but the issue remains relevant. The increase in the post-accident period from the "Chornobyl disaster" has stimulated the greater involvement of the affected territories in intensive economic use. The volume of consumption and procurement of secondary forest use products is increasing. However, the study of their compliance with permissible levels as determined by current legal acts remains relevant. Despite the extensive radioecological

research conducted in the territories affected by the Chornobyl Nuclear Power Plant accident, particularly in the Kamin-Kashyrskyi district of the Volyn region, several unresolved issues regarding radiation safety persist in forest biogeocenoses during the post-accident period. Studies have shown that radioactive elements deposited in forests post-accident, such as radiocesium, tend to reach a quasi-equilibrium within a few years, with the forest retaining these elements over time (Hashimoto et al. 2012, Davydova et al. 2019, Hromyk et al. 2020, Duong et al. 2021), including the presence of contaminated blueberries and mushrooms that are consumed and sold by the local population.

The Chernobyl accident provided a unique opportunity to study the effects of acute and chronic exposure of plants to ionizing radiation, with tens of thousands of hectares of forests experiencing significant radioactive contamination (Arena et al. 2014). Furthermore, research has indicated that the exposure of forests to external γ-radiation over time can lead to increased radiosensitivity in trees due to the accumulation of unrepaired damage (Anspaugh et al. 2002, Real et al. 2004).

Collaborative international efforts, such as those involving the Chernobyl Center's International Radioecology Laboratory, have been instrumental in studying radionuclide accumulation, migration, and the effects of radiation on biological systems in the Chernobyl Exclusion Zone (Bondarkov et al. 2011). Additionally, studies have highlighted the persistence of Chernobyl-derived radionuclides in forest ecosystems, emphasizing the importance of long-term monitoring and understanding the dynamics of radioactive elements in these environments (Fesenko et al. 2006, Koarashi et al. 2014, 2016, Mamyrbayeva et al. 2020. Mandi et al. 2022). The aftermath of the Chernobyl accident has also underscored the need to develop comprehensive strategies for environmental remediation and radiation protection, considering the long-term sustainability of the contaminated areas (Mozaffarian et al. 2011).

While significant progress has been made in understanding the radioecological implications of the Chernobyl accident, ongoing research is essential to address the remaining challenges in ensuring radiation safety in the forest ecosystems affected by the disaster.

However, the results of the research indicate that in the specified territory, and at the present moment, forest products with a significant content of the radionuclide ^{137}Cs, including those exceeding the permissible levels, are to be found (Smith et al. 1993, Handl et al. 2003, Stepanenko et al. 2008, Sekitani et al. 2010, Romanchuk 2012, Harada et al. 2013, Tsubokura et al. 2014, Merz et al. 2015, Nakashima et al. 2015, Koarashi et al. 2020, Shynkarenko et al. 2021, Thiessen et al. 2022).

Therefore, the system "soil-food products of the forest" requires further detailed control, as it is a selective barrier for radionuclides during its passage to the final link of the human trophic chain (Yablokov & Nesterenko 2009, Uwatse et al. 2015, Hromyk & Ilyina 2016, 2017, Grodzynska 2017, Hromyk et al. 2020, Volosovets et al. 2021).

Based on the above, the following research problems were formulated. It is expected that significant levels of radionuclide contamination, particularly ^{137}Cs, persist in the forest products of the Kamin-Kashyrskyi district even 37 years after the Chornobyl disaster, thus the current levels of radiological contamination in forest products from the Kamin-Kashyrskyi district of the Volyn region were investigated. The levels of radioactive contamination in soil and forest products have decreased over time, but at different rates for different ecosystems and product types, thus the distribution of radionuclides (^{137}Cs and ^{90}Sr) in soil and forest products has changed over time since the Chornobyl disaster was first analyzed. Mushrooms exhibit higher bioconcentration factors for ^{137}Cs compared to other forest products such as berries, thus the differences in radionuclide accumulation between various types of forest products (e.g., mushrooms, berries) were investigated. The spatial distribution of radioactive contamination in the study area is heterogeneous, with some areas showing significantly higher levels of contamination than others, thus we investigated to what extent do current levels of contamination in forest products pose a risk to human health. A significant proportion of forest

products in the study area still exceed permissible levels of radionuclide contamination, posing potential health risks to consumers, thus we looked into how effective natural decontamination processes have been in reducing radioactive contamination in forest ecosystems. Natural decontamination processes have had a measurable but limited effect on reducing radioactive contamination in forest ecosystems, thus we analyzed to what extent natural decontamination processes have contributed to the reduction of radioactive contamination in the forest ecosystems of the Kamin-Kashyrskyi district since the Chornobyl disaster. The current levels of radionuclide contamination in forest products necessitate ongoing monitoring and the implementation of preventive measures to protect public health, thus the current levels of radionuclide contamination in forest products from the Kamin-Kashyrskyi district, and how these levels compare to established permissible limits for human consumption were investigated.

These research problems pose a structured framework for investigating the long-term impacts of radioactive contamination in forest ecosystems following the Chornobyl disaster.

MATERIALS AND METHODS

The purpose of the research was to carry out a comprehensive analysis of the dynamics of the content of radionuclide ^{137}Cs in the system "soil-food products of the forest", in particular, forest berries and mushrooms, as a significant source of the formation of the internal radiation dose of the population of the research area.

The starting materials of the research were the results of the authors' own observations (participation in expeditions for the purpose of collecting, processing and analyzing information), scientific publications, as well as presented materials of the Laboratory of Ecological Land Safety and Product Quality of the Volyn Branch of the State Institution "Institute of Soil Protection of Ukraine" (Lutsk). The objects of research were Wild Bilberry (*Vaccinium myrtillus*), Blackberries (*Rubus* L.), Wild Strawberries (*Fragaria vesca* L.), Raspberry (*Rubus idaeus* L.), together with *Cantharellus cibarius*, *Boletus edulis*, and *Leccinum scabrum* mushrooms, sampled in years 2020–2022. The total number of samples was 729 dried and 975 fresh mushrooms and berries samples for 2020, 1,154 dried and 886 fresh samples for 2021, and 870 dried and 896 fresh samples for 2022.

Determination of the volume and specific activity of radionuclides ^{137}Cs and ^{90}Sr was carried out using AI-1024-95 and SEG-05 gamma spectrometers, energy spectrometer of gamma radiation and beta radiation SE-GB-01 AKP-63(Г)-70(B). The results of the internal 10% control over the accuracy of the spectrometric work showed that the main relative error of the measurements does not exceed 7%, which is within acceptable limits.

RESULTS AND DISCUSSION

As the result of the accident at the Chornobyl Nuclear Power Plant, about 3.5 million hectares of forest were exposed to radioactive contamination. In the Volyn, Chernihiv, Cherkasy, Vinnytsia and Sumy regions, the share of radioactively contaminated forests was about 20%. The total area of the territory of the Volyn region with a density of ^{137}Cs pollution from 37 to 185 kBq/m^2 was about 60,000 ha, including about 12,000 ha of agricultural land and 27,600 ha of forests (Merlenko & Zinchuk 2010, Ilyin & Hromyk 2016). In the studied territory of the Kamin-Kashyrskyi district, the forest cover is about 50.2% (Regional report… 2023).

According to the radiological survey carried out in 1991–1993 on an area of more than 500,000 ha, contamination with radionuclides ^{137}Cs and ^{90}Sr was detected on 181,500 ha. The vast majority of soils had a density of ^{137}Cs from 3.7 to 37 kBq/m^2 and amounted to 169,500 ha. The area of the region's land, including agricultural land with a pollution density of more than 37 kBq/m^2, was 12,000 ha (6.6%). The largest number of such lands was found in the Manevychi district – 6,247 ha, Kamin-Kashyrskyi district – 3,832, and Lyubeshiv district– 2,185 ha (after the change of the boundaries of administrative-territorial formations in 2015–2020, these districts were combined into the

Kamin-Kashyrskyi district, which is the subject of this research). For the content of ^{90}Sr, the territories of the Volyn region with a pollution density of up to 0.74 kBq/m^2 – 106,040 ha were counted as surveyed; 0.74–5.55 kBq/m^2 – 75,130 ha; 5.55–111 kBq/m^2 – 308 ha (Ilyin & Hromyk 2016).

According to Pryster (2005), self-cleaning of the territory occurred in the post-accident period due to the natural decay of radionuclides. In 2004, soil monitoring at the control sites of 20 settlements in the contaminated zone, where the passport dose exceeded 1.5 mSv/year, revealed a decrease in the activity of radionuclides in the soil by 15.4–33.3%.

However, monitoring studies have established that despite the relatively low density of soil contamination, quite high levels of radionuclides entering plant products were observed. Among them, significant bioconcentration factors (hereafter referred to as BCF) of ^{90}Sr in berries of forest strawberry (BCF up to 14), raspberry (BCF up to 0.91), and blueberries – BCF up to 0.81 were established. Mushrooms are characterized by a significant excess of BCF ^{137}Cs relative to BCF ^{90}Sr, which can reach 90–400 times. Also, it was established that more than 50% of the cesium radionuclide stock of the entire biota of forest biogeocenoses can accumulate in mushrooms (Grodzynska 2017).

The most detailed radiological studies of soil cover with an accuracy of up to 50 ha were carried out in 2006–2008 (Table 1). According to their results, it was established that of the surveyed 88,682 ha, the levels of ^{137}Cs contamination were: up to 18.5 kBq/m^2 – 93.2% of the area (82,679 ha), 18.5–37 kBq/m^2 – 6.5% (5,774 ha) and more than 37 kBq/m^2 – 0.3% (229 ha) (Regional report… 2023). For ^{90}Sr: up to 0.74 kBq/m^2 – 95.8% (84,978 ha), 0.74–5.55 kBq/m^2 – 4.2% (3,704 ha).

Surveys in 2008 established that there was a decrease in areas with a soil contamination density of more than 37 kBq/m^2 in terms of ^{137}Cs content during the post-accident period – from 11,960 ha in the early 1990s to 229 ha, and in terms of ^{90}Sr content above 0.74 kBq/m^2 – from 75,460 ha to 3,704 ha. Autorehabilitation processes played a significant role in this, particularly wind and water transfer of radionuclides, biological removal with biomass of plants alienated from a certain territory, processes of cleaning the root layer of the soil from radionuclides as a result of vertical migration in the soil profile due to diffusion and convective transfer with water flow and processes of transformation of radionuclide binding forms with the soil, during which their migration ability and biological availability to plants decrease (Pryster 2005, Razanov et al. 2023). No further detailed studies of the soils of the specified territory were conducted.

According to research conducted in 2004, relatively high values of the radionuclide ^{137}Cs were found in a number of food samples, as well as those exceeding the permissible levels (hereinafter – PL) in the by-products of forest use – wild berries and mushrooms. In particular, in the adjacent forest areas of the villages of Kostyukhnivka, Vovchets'k, Kolodiyi, Novi Pidtsarevichi, Nabruska, Lyshnivka, Severynivka, the pollution levels were: in fresh berries – 180 Bq/kg, fresh mushrooms – 925 Bq/kg (Boyko et al. 2017).

Table 1
The content of radionuclides ^{137}Cs and ^{90}Sr in the soils of the polluted zone of the Volyn region (acc. to the data of the Volyn branch of the Protection Institute of Ukraine)

No	Area	Surveyed area [ha]	Pollution density [kBq/m^2]				
			^{137}Cs			^{90}Sr	
			up to 18.5	18.5–37.0	37.0–185.0	up to 0.74	0.74–5.55
1	Manevytskyi	33,640	28,957	4,553	130	32,432	1,208
2	Lyubeshivskyi	25,925	25,101	751	73	24,265	1,660
3	Kamin-Kashyrskyi	29,117	28,621	470	26	28,281	836
Together in the polluted area		88,682	82,679	5,774	229	84,978	3,704

In 2005, studies of the content of ^{137}Cs in dried mushrooms collected near the settlements of the Kamin-Kashyrskyi district revealed a significant excess of the PL contamination levels of the radionuclide (PL-2500 Bq/kg dw), namely Kolodiyi – by 9.3 times (it was 23,300 Bq/kg), Serkhiv – 5.1 times (12,800 Bq/kg), 4.2 times (10,600 Bq/kg) and 1.5 times (3,670 Bq/kg), Ivanomysl – 4.3 times (10,800 Bq/kg), Lobna – 2.7 times (6,850 Bq/kg), 2.6 times (6,540 Bq/kg) and 1.6 times (4,020 Bq/kg), Beef – 2.4 times (6,040 Bq/kg), Zahorivka – 2.2 times (5,500 Bq/kg), Prylisne – 1.5 times (3,760 Bq/kg), Rudka Chervyns'ka and Rakiv Lis – 1.3 times, respectively – 3,380 Bq/kg and 3,240 Bq/kg, Zalissya – 1.2 times (3,010 Bq/kg), Okonsk and Pishchane – 1.2 times (2,920 Bq/kg and 2,910 Bq/kg). The marked PL excess determined the possibility of the internal irradiation of the local population with long-lived radionuclide ^{137}Cs. According to Boyko et al. (2017), forest products are a significant source of radionuclides entering the human body (for the local population – up to 29.8%), including at the expense of dried mushrooms, because when they are dried, the volume decreases significantly, and the concentration of the radionuclide increases (Table 2, Fig. 1).

Fresh mushrooms and berries are also a significant part of the diet of the population of the studied area. In addition, a significant amount of them is sold through a network of procurement points and later used for cooking in restaurants. Radioecological studies for the period 2014–2017 show that in 2014 the maximum level of food contamination was 704 Bq/kg, 2015 – 68 Bq/kg, 2016 – 190 Bq/kg, and 2017 – 204 Bq/kg (at PL – 500 Bq/kg) (Regional report… 2022).

The results of the 2020–2022 research confirm the presence of excesses of PL activity of ^{137}Cs in products of forest origin in 187 samples, which is 3.4% of their total number, with 729 dried and 975 fresh mushrooms and berries samples for 2020, 1,154 dried and 886 fresh samples for 2021, and 870 dried and 896 fresh samples for 2022.

During this period, the maximum contamination levels in dried mushrooms and berries ranged from 7,370 Bq/kg to 8,400 Bq/kg and were detected in 1.9% in 2020, 2.4% in 2021, and 1.8% in 2022.

Table 2

Results of studies of the specific activity of ^{137}Cs in dried mushrooms of the study area of the Volyn region (acc. to the data of the Volyn branch of the Soils Protection Institute of Ukraine)

Settlement	Number of samples	Specific activity, ^{137}Cs [Bq/kg]
Vydrychi	1	457
Vil	1	6,040
Voyehoshcha	1	3,110
Haluziya	1	2,270
	2	18,300
	3	1,670
Hirky	1	1,020
Horodok	1	1,310
Zahorivka	1	5,500
Zalissya	1	2,030
	2	3,010
	3	1,180
Zarika	1	570
Ivanomysl'	1	10,800
	2	2,140
Kolodiyi	1	23,300
Lisove	1	2,180
Lobna	1	6,850
	2	4,020
	3	6,540
	4	1,980
Nichohivka	1	2,430
Novi Chervyshcha	1	2,690
	2	6,710
Okons'k	1	2,920
	2	1,680
Osivtsi	1	152
	2	667
Pischane	1	2,910
Prylisne	1	2,160
	2	3,760
Rakiv Lis	1	3,240
Rudka Chervyns'ka	1	3,380
	2	2,150
Serhiv	1	10,600
	2	12,800
	3	3,670
PL (dried mushrooms)		2,500

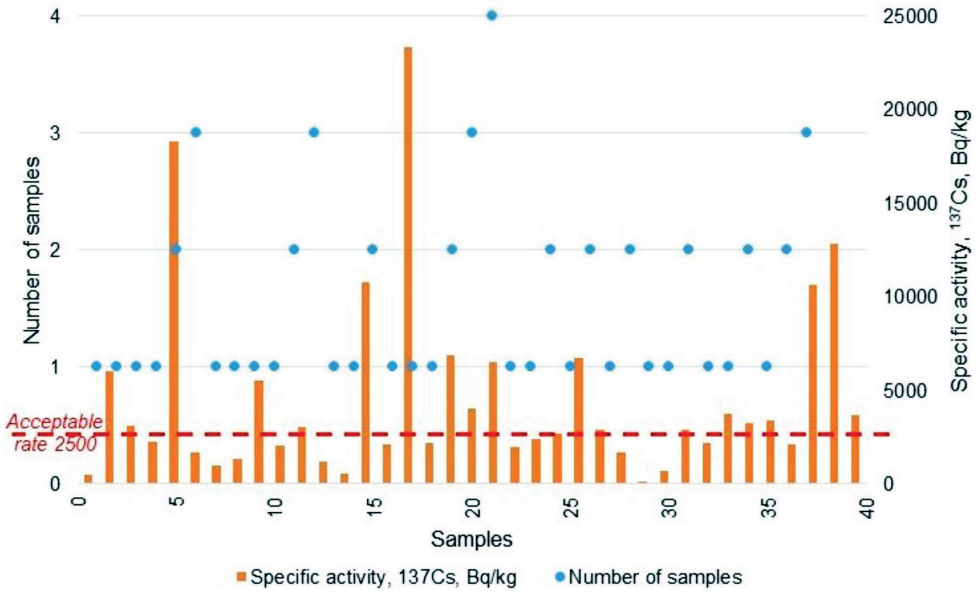

Fig. 1. *Results of studies of the specific activity of ^{137}Cs in dried mushrooms of the study area of the Volyn region*

In fresh mushrooms and berries, an excess of PL of 4.1%, 3.1% and 6.8% was found during the specified period (2020, 2021 and 2022). At the same time, the maximum levels of radiological contamination were 2,430 Bq/kg, 1,270 Bq/kg and 3,005 Bq/kg, respectively. The relative PL excesses were 3.4 times for dried mushrooms and berries, and 6 times for fresh ones (Table 3).

Based on the presented results of radiological research carried out in the period 1991–2022, it was established that despite the decrease in the density of radioactive contamination of the territories of the Kamin-Kashyrskyi district of the Volyn region, the reduction of the volumes of control of the level of radiological contamination of forest berries and mushrooms by 1.8–2.4% of dried products and 3.1–6.8% of fresh products recorded an excess of PL for ^{137}Cs. Data on the content and dynamics of ^{90}Sr in by-products of forest use have not been found, however, given its presence in the soils of forest ecosystems, there is a possibility of its participation in the formation of the total dose load on the population.

Data on the content and dynamics of ^{90}Sr in by-products of forest use have not been found, but given its presence in the soils of forest ecosystems, there is a possibility of its participation in the formation of the total dose load on the population.

Table 3

Dynamics of the specific activity of ^{137}Cs in products of forest origin in the studied territory of the Volyn region (acc. to the data Ministry of Environmental Protection and Natural Resources of Ukraine, 2022)

Product name	Number of tested samples			Number of excesses, PL			The maximum levels of pollution have been detected [Bq/kg]			Permissible level of pollution acc. to PL-2006 [Bq/kg]
	2020	2021	2022	2020	2021	2022	2020	2021	2022	
Dried mushrooms and berries	729	1,154	870	14	28	16	8,400	8,080	7,370	2,500
Fresh mushrooms and wild berries	975	886	896	40	28	61	2,430	1,270	3,005	500

Thus, the results of this study indicate the existence of a problem of radiological contamination of forest products from secondary forest use, which requires further strengthening of radiological control.

Based on the data and analysis presented in the article, we can formulate the following summary of the analyzed research problems.

1. The analysis of the results of studies of the levels of radionuclide contamination of the soil cover of the Kamin-Kashyrskyi district of the Volyn region for the period 1991–2008 shows trends in the direction of improving the radiological situation in the territories affected by the accident at the Chornobyl nuclear power plant. Long-term contamination persistence: The study confirmed that even 37 years after the Chornobyl disaster, significant levels of radionuclides, particularly ^{137}Cs, were still present in forest products. This supports the hypothesis of long-term radioactive contamination in the affected areas.
2. Despite the improvement of the radiological situation as a whole, the presence of the problem of radiological contamination of ^{137}Cs by products of secondary forest use in the studied territory has been established. For the period of 2020–2022, an excess of PL activity of ^{137}Cs was detected in 187 samples of forest products, which is 3.4% of their total number, including 1.8–2.4% of dried samples and 3.1–6.8% of samples of fresh products, and the maximum levels of PL exceedance were 3.4 times for dried mushrooms and berries, and 6 times for fresh ones. Gradual decrease in contamination levels: While a general downward trend in soil contamination levels was observed over time, forest products continued to show elevated levels of radionuclides. This partially confirms the hypothesis of decreasing contamination levels but highlights the slower decontamination process in forest ecosystems.
3. The presented results indicate the presence of risks of internal radiation of the population of the studied territory due to the consumption of wild berries and mushrooms, which requires strengthening of radiological control during the period of harvesting and sale of products, as well as expanding the network of radiological monitoring in order to take timely anti-radiation measures. Due to the significant reduction in contamination, taking into account the amount of consumption of the tested products, the risk appears to be reduced. Spatial variability of contamination. The research revealed significant differences in contamination levels between different areas within the studied region, confirming the hypothesis of uneven distribution of radioactive contamination.
4. Higher radionuclide accumulation in mushrooms. The study demonstrated that mushrooms had significantly higher bioconcentration factors for ^{137}Cs compared to ^{90}Sr, confirming the hypothesis of greater radionuclide accumulation in fungi.
5. Ongoing monitoring necessity. The persistent elevated levels of radionuclides in forest products, often exceeding permissible levels, underscore the need for continued radiological monitoring. This confirms the hypothesis regarding the necessity of long-term surveillance.
6. Limited effectiveness of natural decontamination. While ome natural processes of radionuclide deconcentration were observed, their effects were limited, especially in forest products. This partially supports the hypothesis on the effectiveness of self-cleaning processes but highlights their insufficiency in forest ecosystems.
7. Public health implications. The study indicated a potential risk of internal radiation exposure for the local population due to the consumption of contaminated forest products, emphasizing the ongoing public health concern. Need for targeted preventive measures: Given the persistent contamination in forest products, there is a clear need for the continued implementation of targeted preventive measures and public awareness campaigns.
8. Importance of ecosystem-specific approach. The research highlighted the unique dynamics of radionuclide behavior in forest ecosystems, suggesting the need for ecosystem-specific approaches in long-term radioecological management strategies. These findings collectively emphasize the enduring impact of the Chornobyl disaster on forest ecosystems and the need for ongoing vigilance, monitoring, and adaptive management strategies to ensure the safety of forest products and to protect public health in the affected regions.

CONCLUSIONS

Studies showed that significant levels of radionuclides, especially ^{137}Cs, were still detected in forest products even 37 years after the Chornobyl disaster. A general downward trend in soil contamination levels was observed, but forest products still showed elevated levels of radionuclides. Our research data confirmed the significant differences in contamination levels between different investigated areas. Studies showed that mushrooms were characterized by significantly higher bioconcentration factors for ^{137}Cs compared to ^{90}Sr. Research results indicate the need to continue the radiological monitoring of forest products due to the persistent risk to human health. Considering the effectiveness of self-cleaning processes, it was revealed that some natural processes of radionuclide deconcentration were observed, but their effects are limited, especially in forest products. The research confirmed the long-term impact of the Chornobyl disaster on forest ecosystems and the products derived from them. Despite the passage of time, the problem of radioactive contamination remains relevant and requires further monitoring and preventive actions.

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