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N. E. Zarubina[1,*], O. S. Burdo[1], L. P. Ponomarenko[2], O. V. Shatrova[3]

[1] *Institute for Nuclear Research, National Academy of Sciences of Ukraine, Kyiv, Ukraine*
[2] *National Technical University of Ukraine "Igor Sikorsky Kyiv Polytechnic Institute", Kyiv, Ukraine*
[3] *SE "UKRMETRTESTSTANDARD", Kyiv, Ukraine*

*Corresponding author: nataliia.zarubina@gmail.com

TWO STAGES IN THE ACCUMULATION OF ^{137}Cs BY MUSHROOM *SUILLUS LUTEUS* AFTER THE CHORNOBYL ACCIDENT

Studies of the ^{137}Cs content in fruit bodies of *Suillus luteus* in the territory of the Chornobyl exclusion zone and Kyiv region outside the zone were carried out during the period 1986 - 2020. It was found that the dynamics of ^{137}Cs activity in the mushroom can be described as a two-stage process. The first stage since 1986 was characterized by the annual increase in levels of specific activity of ^{137}Cs for the following 10 - 12 yrs. During the second stage, there has been a gradual decrease in concentrations of ^{137}Cs. The ecological half-life of ^{137}Cs in the mushroom at the second stage differs for different sampling sites. Its minimum values were noted at Yaniv and Novo-Shepelychy sampling sites inside the exclusion zone. The maximum of ^{137}Cs ecological half-life in *Suillus luteus* was observed on the Rzhyshchiv sampling site, which is the most remote from the Chornobyl Nuclear Power Plant outside the exclusion zone.

Keywords: mushroom, *Suillus luteus*, ^{137}Cs, Chornobyl (Chernobyl) NPP accident, Chornobyl (Chernobyl) exclusion zone, two stages.

Since the Chornobyl accident on 26 April 1986, a lot of publications devoted to the study of the accumulation of artificial radionuclides by mushrooms in the forests of the Northern Hemisphere have appeared in the scientific literature.

Studies [1, 2] carried out in Europe indicate an increase in the content of radionuclides, and in particular of two Cs (^{137}Cs and ^{134}Cs) radioisotopes in mushrooms since 1986, compared to the pre-accident values. Thus, in *Paxillus involutus* sampled in Austria [2], ^{137}Cs concentrations increased 5 - 10 times; in *Suillus variegatus* – 60 - 130 times; in *Lactarius rufus* – 20 - 40 times higher. In Poland, the content of this radionuclide increased by a factor of 10 in fungi of different species [3].

In the initial 3 yrs after the accident, the average ^{137}Cs content in mushrooms in Europe was higher than in mosses and lichens [4] and 5 to 270 times higher than in vascular plants [5].

Mushrooms belonging to the ecological group of symbiotrophs are the most contaminated with ^{137}Cs [6, 7]. However, the content of radiocaesium in symbiotic mushrooms growing within a short distance from each other can vary significantly (by the order of magnitude) [8]. This is due to one of the main factors affecting the accumulation of ^{137}Cs by mycorrhizal mushrooms, the deep localization of the main part of the mycelium of each species in the underlying soil [9]. An order by the amount of ^{137}Cs accumulated by various forest ecosystem objects following the Chornobyl accident is as follows: mycorrhizal mushrooms > mosses and lichens > higher plants [4, 5, 10, 11].

The intensity of the absorption of Cs isotopes from the soil by mushrooms is higher than in plant biota of the forest, and they accumulate the radiocaesium more intensively than natural isotope ^{40}K [12 - 14]. Studies in Poland, Italy, and Finland have shown that mushrooms almost do not accumulate ^{90}Sr and transuranic elements, as well as other artificial radionuclides [8, 15, 16]. Our studies found that in the territory of the Kyiv region (Tolokun) radionuclides were present in *Boletus edulis*: ^{137}Cs – 39 ± 17, ^{131}I – 115 ± ± 45, and the pair ^{140}Ba-^{140}La – 229 ± 37 Bq/kg of fresh weight in June 1986. In *Cantharellus cibarius* and *Xerocomus chrisenteron* in the territory of the Chornobyl NPP exclusion zone contained ^{134}Cs, ^{106}Ru, ^{103}Ru, ^{144}Ce, ^{141}Ce, ^{95}Zr, ^{95}Nb, ^{140}Ba-^{140}La in July 1986, i.e., almost all short-lived radionuclides of the Chornobyl emissions [17].

The problem of accumulation of radiocaesium by mushrooms again attracted the attention of scientists after the accident at the Fukushima Dai-ichi NPP, Japan in 2011, resulting in contamination of large forest areas by artificial radionuclides. Several publications on mushrooms pollution after the accident have appeared in the scientific literature [18 - 20].

Analyzing the consequences of these two largest nuclear accidents with the dissipation of radionuclides into the environment the following should be noted: firstly, when artificial radionuclides enter the environment, they necessarily pollute forests to one degree or another; secondly, in the forest ecosystems mushrooms are one of the maximum accumulators of biologically significant, long-lived radionuclide ^{137}Cs; thirdly, the fruit bodies of mushrooms are one of the links of the trophic chain, leading to man.

The aim of our paper is research of ^{137}Cs accumulation in *Suillus luteus* (L.) Gray on the territory of the Chornobyl exclusion zone and the Kyiv region outside the zone after the accident and in a perspective.

This species of mushrooms belongs to the class of basidiomycetes, a family of boletes (Boletaceae). *S. luteus* is an obligate symbiotroph and forms mycorrhiza with *Pinus sylvestris* L. on the territory of Ukrainian Polissia [21]. The main part of mycelium of this species is localized in the near-surface 0 - 5 cm soil layer [22, 23].

1. Materials and methods

1.1. Sampling sites

We studied ^{137}Cs accumulation by mushrooms on the territory of the exclusion zone of Chornobyl Nuclear Power Plant (ChNPP) and the Kyiv region outside the exclusion zone. Sampling sites are located at different distances and directions from ChNPP (Figs. 1 and 2).

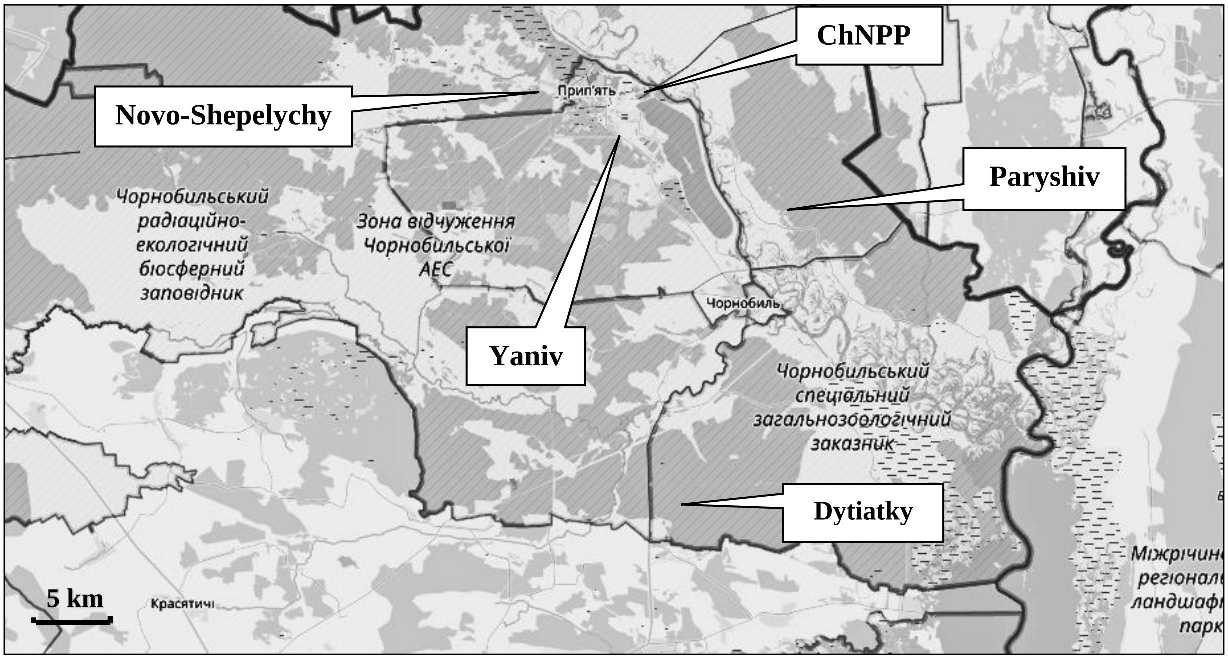

Fig. 1. The sampling sites within the Chornobyl exclusion zone.

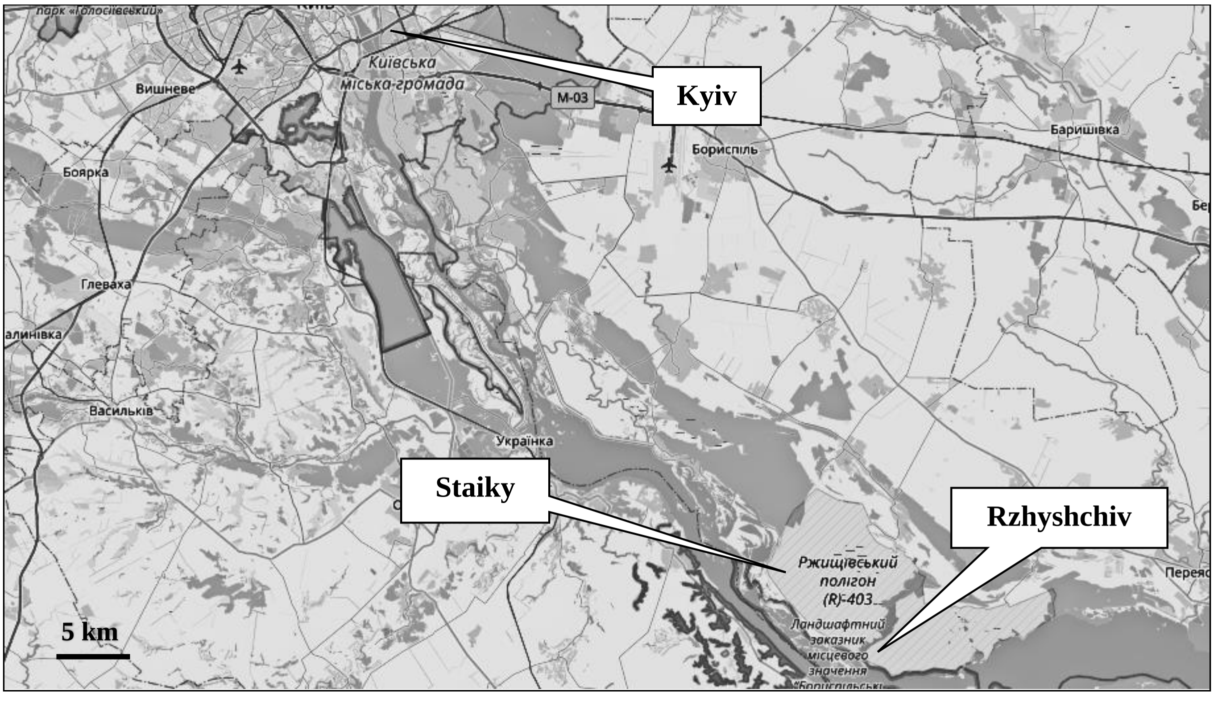

Fig. 2. Sampling sites at the territory of Kyiv region outside the zone.

The sampling sites within the Chornobyl exclusion zone have the following coordinates: Yaniv (30˚04'01.51"E, 51˚23'22.18"N), Novo-Shepelychy (30˚00'32.83"E, 51˚25'04.77"N), Paryshiv (51˚17'57.54"E, 30˚18'17.43"N) and Dytiatky (30˚07'21.83"E, 51˚07'13.37"N).

The coordinates of the sampling site within the Kyiv region are as follows: Staiky (30˚59'45.21"E, 50˚02'37.76"N) and Rzhyshchiv (31˚02'01.75"E, 50˚01'40.86"N).

Sampling of *S. luteus* fruit bodies was carried out: Dytiatky – from 1986 till 2020; Staiky – from 1986 till 2012; Novo-Shepelychy – from 1988 till 2006; Rzhyshchiv – from 1994 till 2008; Yaniv – from 1997 till 2008; Paryshiv – from 1999 till 2020. The size of each sampling sites is approximately 100 m^2. The soil at the sampling sites belongs to the sod-podzol type. Scots pine (*P. sylvestris*) prevails among wood vegetation on the territory of the studied sampling sites.

1.2. Sampling of fruit bodies of fungi

Samples of complete fruit bodies (caps and stipes) of *S. luteus* were collected during the period of the massive occurrence.

The fruit bodies of average size, with no visible external damage were collected from areas of up to 100 m^2. The samples were cleaned from surface contaminations and homogenized using a blender. Mixed mushrooms sample consisted of 3 - 7 whole fruit bodies. Data of the measurements of the ^{137}Cs specific activity and measurement errors are given in this article for one mixed mushrooms sample in one moment of the time on each sampling site.

1.3. Radiometry

^{137}Cs specific activity measurements were performed employing a CANBERRA gamma-spectrometric set-up based on coaxial highly pure HPGe semiconductor detector, model GC6020.

The detection unit was covered with 100-mm lead protection allowing to carry out effective measurements of samples with a comparatively low radionuclide specific activity.

The measurement time was from 600 to 14,400 s depending on the specific activity of the radionuclide. The measurement errors of this series of samples did not exceed 10 % and, as a rule, were within the limits of 3 - 5 % of the radionuclide activity.

In this article, the ^{137}Cs specific activity in mushrooms is given in Bq/kg of fresh weight.

A detailed description of the materials and methods is given [9].

Statistical software packages “Statistica” and “Excel” were used for mathematical data processing.

2. Results and discussion

2.1. Changes in ^{137}Cs content in *S. luteus* after the Chornobyl accident can be divided into two stages (Figs. 3 - 5). The first stage was characterized by an annual increase in the content of radiocaesium in the fruit bodies of this species of mushrooms and lasted from 1986 till 1996 - 1998 (depending on a sampling location).

Fig. 3. The specific activity of ^{137}Cs in *S. luteus*, sampling site Novo-Shepelychy, Bq/kg fresh weight. The dashed line shows the calculated line of ^{137}Cs ecological half-life.

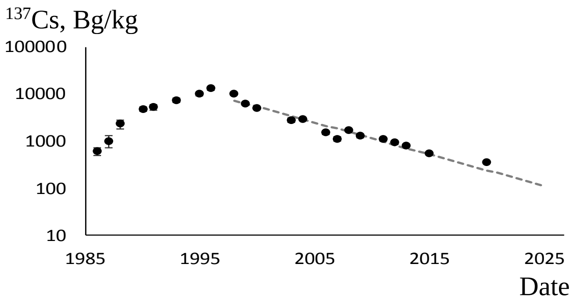

Fig. 4. The specific activity of ^{137}Cs in *S. luteus*, sampling site Dytiatky, Bq/kg fresh weight. The dashed line shows the calculated line of ^{137}Cs ecological half-life.

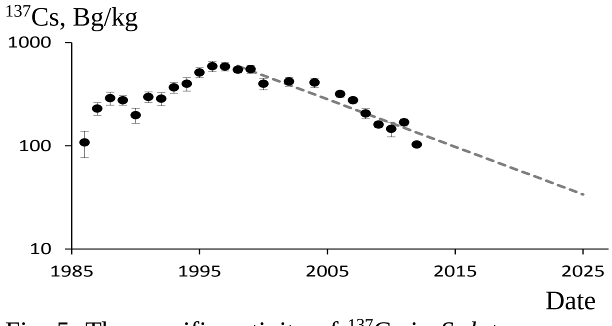

Fig. 5. The specific activity of ^{137}Cs in *S. luteus*, sampling site Staiky, Bq/kg fresh weight. The dashed line shows the calculated line of ^{137}Cs ecological half-life.

During this stage, the specific activity of ^{137}Cs in *S. luteus* increased by approximately 10 times. The maximum levels of ^{137}Cs content in this species of mushrooms were observed in 1996 - 1998.

The content of ^{137}Cs in fungi of one species in Europe after the Chornobyl accident was not constant and changed over time. A decrease in ^{137}Cs specific activity levels in mushrooms, the mycelium of which is in the undecomposed and semi-decomposed layers of forest litter, was observed on the territory of Ukrainian Polissia during 1987 - 1994. Significant changes in the content of radiocaesium in mushrooms, the mycelium of which is located in the decomposed part of the forest litter, were not observed during this period. The increase in the amount of ^{137}Cs accumulated in fruit body, the mycelium of which is located in the upper humified part of the mineral soil, was noted [24]. The increase in ^{137}Cs specific activity in *Imleria badia* was noted in the Chornobyl exclusion zone and Kyiv region from 1986 till 1997 [25]. In Poland, the maximum ^{137}Cs content in *B. edulis* and *I. badia* was observed in 1988 - 1989, in *C. cibarius* in 1991 [26]. In Pomerania (Poland) the highest concentrations of radiocaesium were observed in *B. edulis* in 1999 for the period 1995 to 2015 [27]. The increase in the content of ^{137}Cs in some species of mushrooms (*Hydnum repandum*, *I. badia,* etc.) in the period up to 1997 on the territory of France was reported. The concentration of radiocaesium increased as much as twice and more in fruit bodies from 1992 till 1997 [28]. The research of the radiocaesium content in mushrooms on the territory of Norway made it possible to suppose that the maximum of the content of this radionuclide in different species will be observed in different years [29]. The highest concentrations of ^{137}Cs in various species of mushrooms sampled on the same territory could be noted in different periods (indicated in [30]).

The increase in levels of ^{137}Cs specific activity in *S. luteus* at the first stage was observed not only at the sampling sites contaminated by the fuel component of the fallout [16, 31]. This process was also noted in areas where pollution was caused by precipitation of condensational components of the fallout (see Figs. 3 - 5). It can be supposed that the increase in ^{137}Cs content in *S. luteus* on the territory of the research at this stage could be associated not only with the output of this radionuclide from “hot” particles but also with the gradual rising in its bioavailability during the first 10 to 12 yrs after the accident. Thus, the process of ^{137}Cs involvement from Chornobyl fallout in the biological chains in the forest ecosystems has a considerable duration.

2.2. A decrease in the levels of ^{137}Cs specific activity in *S. luteus* is observed at the second stage (from 1996 - 1998 till now) (see Figs. 3 - 5).

We calculated the ecological half-life of ^{137}Cs for *S. luteus*, sampled at sampling sites with different levels of radiocaesium content in soils [32, 33].

The specific activity of ^{137}Cs in *S. luteus* at the second stage is as follows:

$$A(t) = A_0 \exp\left(-\ln 2 \frac{t}{\tau_{eff}}\right),$$

where A_0 – specific activity of ^{137}Cs at time zero; τ_{eff} signifies the effective half-life, which depends on both the ecological half-life and the decay period.

If the values of the ^{137}Cs concentration convert into a logarithmic scale, we get a straight line *b - at* along which the experimental points are scattered. The parameters of this straight line can be determined using the standard/ordinary method of least squares. Knowing *a* one can determine the effective half-life:

$$\tau_{eff} = \ln 2 / a.$$

In this case, the behavior of the specific activity in time will be approximated by the dependence

$$A(t) = \exp(b)\exp\left(-\ln 2 \frac{t}{\tau_{eff}}\right).$$

Based on proceeding from the known effective half-life τ_{eff} and decay period (half-life period of ^{137}Cs, $\tau_{1/2}$ = 30.2 yrs), it is easy to calculate the ecological half-life - $\vartheta_{1/2}$ from the ratio:

$$\frac{1}{\tau_{1/2}} + \frac{1}{\vartheta_{1/2}} = \frac{1}{\tau_{eff}}.$$

The $\vartheta_{1/2}$ values for sampling sites with different levels of soil pollution are very noticeably different (Table). The minimum value $\vartheta_{1/2}$ was obtained for the Yaniv sampling site (2-km Chornobyl zone) – 3.14 yrs; maximum – at the Rzhyshchiv sampling site (the territory of the Kyiv region, the distance from the Chornobyl nuclear power plant is approximately 160 km) – 11.91 yrs. As an example, the Table shows the values of the specific activity of ^{137}Cs in *S. luteus* and the density of soil contamination at the sampling sites in 2004. In Figs. 3 - 5, the dashed lines show the calculated line of ^{137}Cs ecological half-life for up to 2025 at the sampling site.

In the result of the calculation of the ecological half-life ^{137}Cs it was found that in the territories of the 5-km zone, i.e., in the forest ecosystems with the largest levels of soil pollution, $\vartheta_{1/2}$ ^{137}Cs is the lowest. It means that in the territories with the maximum level of soil pollution, the specific activity of this radionuclide in mushrooms decreases at a greater rate. With the increase in distance from the ChNPP $\vartheta_{1/2}$ ^{137}Cs in *S. luteus* increases as much as several times. A maximum of $\vartheta_{1/2}$ ^{137}Cs was obtained for the Rzhyshchiv sampling site, the most remote from the ChNPP (see Figs. 3 - 5).

The difference in $\vartheta_{1/2}$ ^{137}Cs in areas with different levels of contamination may indicate the existence of a complex mechanism for the removal of cesium by mushrooms. At highly contaminated sampling sites, they more intensively remove the “excess” ^{137}Cs from their body. At the first stage, the intake of ^{137}Cs into the body of mushrooms exceeded the limits of possible removal, since the beginning of the second stage the mechanism of removal proves to manifest itself more.

The value of $\vartheta_{1/2}$; the content of ^{137}Cs in *S. luteus*, equivalent dose rate, 2004

Sampling site (distances and direction from ChNPP	$\vartheta_{1/2}$, yrs	$\Delta\vartheta_{1/2}$, yrs	*A*(2004), Bq/kg	Δ*A*(2004), Bq/kg	Equivalent dose rate, μSv/h, 2004
Inside the exclusion zone					
Yaniv (2 km, to the West)	3.14	0.51	2,700,000	16,900	15.00
Novo-Shepelychy (6 km, to the Westnordwest)	4.14	2.22	48,700	2,300	3.50
Dytiatky (30 km, to the South)	5.37	0.42	2.900	300	0.30
Paryshiv (20 km, to the Southeast)	7.21	0.71	1.810	80	0.35
Outside the exclusion zone					
Staiky (150 km, to the Southsoutheast)	8.34	0.77	410	30	0.20
Rzhyshchiv (160 km, to the Southsoutheast)	11.91	3.79	270	20	0.20

3. Conclusion

Summing up the results of the long-term research of ^{137}Cs accumulation by *S. luteus* carried out on the territory of the Chornobyl exclusion zone and Kyiv region with the different levels of pollution by this radionuclide following the accident at the ChNPP, it was discovered that dynamics of radiocaesium content in mushrooms was represented by a two-stage process. The first stage was characterized by the annual increase in levels of the specific activity of ^{137}Cs in mushrooms and lasted for the initial 10 - 12 yrs, after 1986. At the second stage, there was a gradual decrease in ^{137}Cs concentration in *S. luteus* and this process has been continuing so far.

The calculation of ^{137}Cs ecological half-life showed that on the sampling sites which are in close proximity to the ChNPP (villages Yaniv and Novo-Shepelychy), the rate of decrease concentration of this radionuclide in *S. luteus* at the second stage is much higher than on the sampling sites with the lower level of soil pollution. The cause for the differences in the value of ^{137}Cs ecological half-life on territories with the different levels of soil pollution may be the mechanism of removal of "excess" radiocaesium by mushrooms.

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Н. Є. Зарубіна[1,]*, О. С. Бурдо[1], Л. П. Пономаренко[2], О. В. Шатрова[3]

[1] *Інститут ядерних досліджень НАН України, Київ, Україна*
[2] *Національний технічний університет України*
«Київський політехнічний інститут імені Ігоря Сікорського», Київ, Україна
[3] *ДП «УКРМЕТРТЕСТСТАНДАРТ», Київ, Україна*

*Відповідальний автор: nataliia.zarubina@gmail.com

ДВА ЕТАПИ НАКОПИЧЕННЯ ^{137}Cs ГРИБАМИ *SUILLUS LUTEUS* ПІСЛЯ ЧОРНОБИЛЬСЬКОЇ АВАРІЇ

Дослідження вмісту ^{137}Cs у плодових тілах *Suillus luteus* на території Чорнобильської зони відчуження та Київської області за межами зони проводились у період 1986 - 2020 рр. Установлено, що динаміку активності ^{137}Cs у цьому виді грибів можна описати як двоступеневий процес. Перший етап з 1986 р. характеризувався щорічним збільшенням рівня питомої активності ^{137}Cs протягом перших 10 - 12 років. На другому етапі спостерігалося поступове зниження концентрацій ^{137}Cs. Екологічний період напіввиведення ^{137}Cs у цьому виді грибів на другому етапі відрізняється для різних місць відбору проб. Мінімальні його значення відзначено на місцях відбору проб сіл Янів та Ново-Шепеличі. Максимум екологічного періоду напіввиведення ^{137}Cs у *Suillus luteus* спостерігається на полігоні Ржищів, який є найбільш віддаленим від Чорнобильської АЕС.

Ключові слова: гриби, *Suillus luteus*, ^{137}Cs, аварія на Чорнобильській АЕС, Чорнобильська зона відчуження, два етапи.

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