Species Dependence and Temporal Dynamics of ^{137}Cs Contamination of Mushrooms from Shatsk and Carpathian Natural National Parks

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Abstract—**The results of the research of changes radioactive caesium contamination for some types of mushrooms from the territory of Shatsk (Volyn region) and Carpathian National Natural Parks (ShNNP and CNNP, respectively) in 1114-2012 are presented. Interspecific and intraspecific dependences of ^{137}Cs contamination of mushrooms as well as the factors that affect them were analyzed. It has been noted that despite the fact that soil contamination slowly decreases over time, during the mentioned time there was no clear tendency to reduce radiocaesium contamination in fungal bodies.**

Index Terms—**radionuclide contamination, radiocaesium, specific activity, soil contamination density, transfer factor**

I. Introduction

Investigation of radioactive contamination of mushrooms is very important because wild mushrooms have not only valuable nutritional qualities, but also are specific components of forest biogeocoenoses, as they play an important role in their functioning, in particular – in the migration of radionuclides in the environment [1, 2]. The contribution of mushrooms to the circulation of ^{137}Cs in forest objects several times exceeds the contributions of the tree and grass-bush tier. Mushrooms also are one of the main factors determining the role of litter as a biochemical barrier in the path of vertical migration of radionuclides in forest ecosystems [3, 4] and can serve as bioindicators of radioactive contamination of the environment [3, 5].

Investigating the dynamics of the state of radioactive contamination of environmental objects allows not only to estimate the level of its radioactive contamination at a specific time in a particular place and under specific conditions, but also to predict its changes. The importance of study the radiological status of mushrooms in the Carpathians and Polissya is also largely because these regions are important sources of these foods of human nutrition in Ukraine. According to estimates [3], the contribution of wild mushrooms and berries to the internal irradiation of the population of the region can reach 75-80% of the total dose of internal exposure that it receives from the consumption of all food products.

The intensity of the accumulation of ^{137}Cs in the fetal body of mushrooms plays a decisive role in their belonging to a certain trophic group [3, 4, 6]. Since each type of mushrooms has a certain depth of occurrence of mycelium [1, 7], it is obvious that the accumulation of their fetal bodies of radionuclide will be determined primarily by the presence of the latter in the appropriate layer of the nutrient substrate [7] in an available for absorption form. Mushroom growth conditions, especially the type of soil and climatic factors, are also very significant [1, 2]. There is also a seasonal dependence of the ability to accumulate radiocaesium mushrooms of the same species in the same growth location. Thus, the author [8] argues that during the season, the content of ^{137}Cs in the selected mushrooms of the same species may differ by more than an order of magnitude. There is data on the age-related dependence of the content of radiocaesium in mushrooms - young and old mushrooms of the same species, collected in one place, have different specific activity of radiocaesium, and with the attainment of the fungus, the radionuclide content in its cap and leg is also changed, and in the head it becomes larger [9, 10].

II. Mayerial and Metods

For research, samples of mushrooms and soils on they grow were taken.

Samples to determine the content of radionuclides in the upper 20 cm soil layer were taken annually in the first half of July (in order to reduce the influence of seasonal factors) on the same sites using the "envelope" method and were prepared for analysis according standard technique.

Preparation of selected samples of mushrooms for gamma analysis consisted of sorting the selected material into appropriate vegetative and generative organs (on caps and stipes), their chopping, drying in a drying box at a temperature of 80°C for at least 24 hours, grinding and homogenization.

For gamma-analysis, the required amount of prepared in this way sample, was selected, weighed and placed in a special container in the gamma-gamma spectrometer measuring chamber.

Qualitative and quantitative analysis of the investigated samples was carried out on a gamma spectrometer with semiconductor Ge(Li)-detector DGDK-100B in an accredited measuring laboratory of the Ivan Franko National University of Lviv. Protection from external gamma radiation was carried out by shielding the measuring chamber of the spectrometer with a 10 cm thick lead screen.

The applied measurement technique made it possible, by varying the time of the set of apparatus spectrum, to obtain the values of the calculated activity of radionuclide in the investigated samples (A, Bq/kg) and the density of soil contamination (S_s, Bq/m^2) with an error of not more than ±15%.

Using the values of these values, the coefficients of transition of radionuclide radionuclide from soil to mushroom (transfer factor T_f) were calculated [11]:

$$T_f = A/S_s, \quad (1)$$

where is A – specific ^{137}Cs activity in the mushroom.

III. Results and discussion

The results of our studies have shown that the contamination of the soils of the Carpathians and Polissya has a mosaic character. The ^{137}Cs contamination of the soils of the Chornogora Range (CNNP) changes almost 5 times - within the range of 5-33 kBq/m^2, depending on the geographical position of the site of selection and its altitude and is more larger than ones for the radionuclide content in the soils of the ShNNP (2.6-8 kBq/m^2) [12]. This situation is evidently due to the higher density of initial deposition of radioactive clouds of this radionuclide in mountainous areas compared with its deposition on the territory of ShNPP in the post-Chornobyl period. In addition, the natural conditions and the related peculiarities of radionuclide migration led to a certain difference in the distribution of ^{137}Cs in the soils of these regions with depth. An important aspect is the kind of soil. Realy, in the Carpathians there are the predominantly brown soils and the main stock of available radionuclides in soils is in their upper 5-10 cm layers. A similar type of change in the distribution of radiocaesium for the soil in depth occurs in gray forest and sod-podzolized soils of the ShNNP only, and there is an almost smooth decreasing of the amount of radionuclide with depth for sandy soils of this park [12].

The research conducted by us have shown that in all years there is a significantly higher content of radionuclide in mushrooms in ShNNP in comparison with CNNP, despite the fact that, on the contrary, the density of soils contamination with radiocaesium in Chornogora is more than 8 times higher. Observed significant fluctuations in the values of ^{137}Cs contamination in mushrooms are due, obviously, to different growth conditions, since mushrooms were selected at different parts of the parks and characterize the accumulation precisely in relation to these territories. It is known that, depending on the conditions of growth (in particular, the type of soil), the amount of accumulated radionuclide in the fungal bodies of the fungus can vary in several dozen times [10]. At the same time, there is no clear tendency to noticeably decrease over time the content of ^{137}Cs in the fungal bodies of mushrooms, which is consistent with known literature data [1]. While held earlier [13] studies have shown a tendency to reduce the radionuclide content in the upper 20-cm soil layer in SHNPP over the years, is described as $0.16e^{-0.0251t}$ ($R^2 = 0.73$).

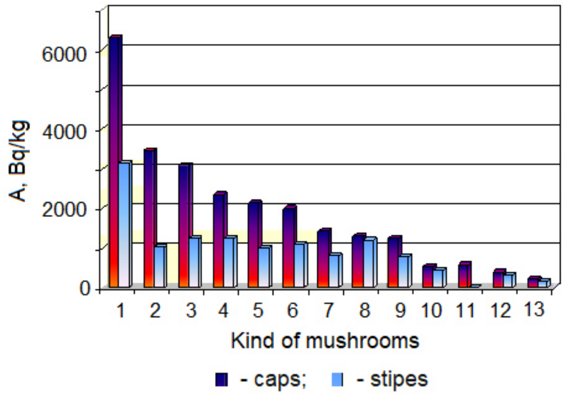

Fig. 1. ^{137}Cs contamination in mushrooms of various species for ShNNP (sampling time July 2001) :.

1 – *Boletus felleus;* 2 – *Russula rosea;* 3 – *Russula heterophylla;* 4 – *Russula ochroleuca*; 5 – *Russula claroflava*; 6 – *Russula decolorans*; 7 – *Russula badia*; 8 – *Cantharellus cibarius*; 1 – *Russula paludosa*; 10 – *Russula cyanoxaniha*; 11 – *Scleroderma*; 12 – *Boletus edulis*; 13 – *Boletus subtomentosus*.

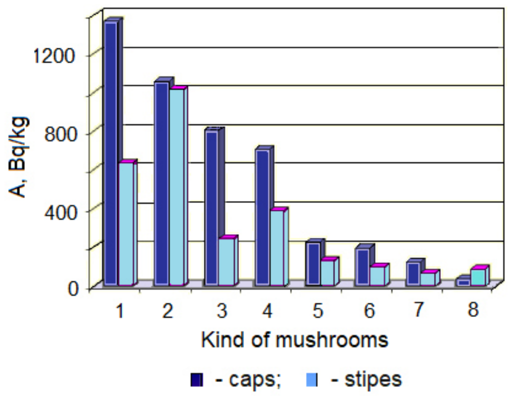

Fig. 2. ^{137}Cs contamination in mushrooms of various species for CNNP (sampling time August 2001):

1 – *Hydnum repandum*; 2 – *Boletus badius*; 3 – *Boletus edulis*; 4 – *Hidnum erinaceum*; 5 – *Boletus aurantiacus*; 6 – *Russula vesca*; 7 – *Boletus satanas*; 8 – *Armillaria mellea.*

Dependence of ^{137}Cs contamination in mushrooms of various kinds which grown on the territory of ShNNP (sampling time - July 2001) is shown in Fig. 1 whereas on the territory of CNNP (sampling time - August 2001) – in Fig. 2. As can be seen, in general, the contamination level of mushrooms in the ShNNP is higher ones in the CNNP, despite the difference in the contamination of their soils. Also, we can see an excess of 137Cs contaminants in the caps above the stipes for all the examined fungi.

It is known that the determining factor in the formation of radioactive contamination is the depth of lying of the mycelium of each type of fungi, as well as the fixation of radionuclide in soil complexes, which is much higher in brown soils than in the acidic soils of Polissya [3]. It is obvious that the difference between the amount of radiocaesium soil contamination and varying availability of radionuclide for assimilation causes the difference in the contamination of mushrooms in the investigated areas mainly.

Intraspecific dependence of the ability to accumulate radiocaesium by mushrooms was studied on the example of the most widespread in the forests of Ukraine representatives of the *Russulaaseae* genus, sampled on the territory of ShNNP in July 2001. The ^{137}Cs specific activity in the examined caps and stipes of the investigated mushrooms genus *Russulaaseae* is shown in Fig. 3.

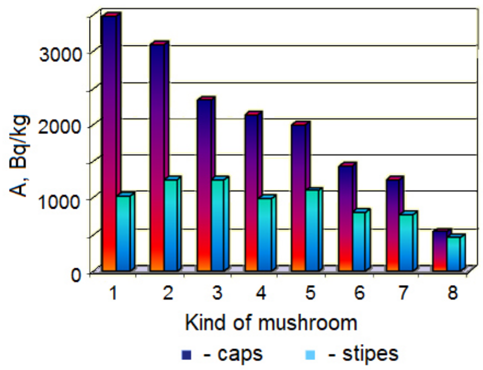

Fig. 3. Special activity of ^{137}Cs in different kinds of mushrooms genus *Russulaaseae* from ShNNP (in dry weight; sampling time July 2001):

1 – *Russula rosea*; 2 – *Russula heterophylla*; 3 – *Russula ochroleuca*; 4 – *Russula claro-flava*; 5 – *Russula decolorans*; 6 – *Russula badia*; 7 – *Russula paludosa*; 8 – *Russula cyanoxaniha.*

Significant differences in the ability to accumulate radionuclide by representatives of one family are no due to the conditions of growth, because mushrooms for investigations were collected at the same section of a forest with practically identical soil cover in the northern part of the shore of Pisochne Lake. Obviously, one of the reasons of this is the difference in the depth of mycelia placing for the certain species of mushrooms in the family - the most contaminated under the same conditions of growth will be those types of mushrooms, whose mycelium are located in a layer of soil with the greatest amount of radionuclide, available for it assimilation. Probably, these particularities in the ability to accumulate radiocaesium by certain types of fungi have largely determined the differences in the ranking of fungi for their ability to accumulate ^{137}Cs, which are noted in the works of various authors [1-3, 6-8]. Probably, it is precisely these particularities in the ability to accumulate radiocaesium by certain kinds of fungi have largely determined the differences in the ranking of fungi for their ability to accumulate 137Cs, which are noted in the works of various authors [1-3, 6-8].

In all types of studied mushrooms (with the exception of mushrooms *Armillaria mellea*) there was an excess of ^{137}Cs content in the caps in comparison with their stipes, which is in a good agree with [9, 10].

From the point of view of estimating the contribution of mushrooms to the cycle of radioactive elements in forest biocenoses, and especially in the additional dose of radiation of people, it is important not only to have data on their contamination at the given time, but also about its change over time.

Changes of ^{137}Cs (A, Bq/kg) content in some of the most popular edible species of wild mushrooms (and most commonly used in food) from SHNPP in 2007-2012 are shown in Fig. 4 (dry weight; sampling of mushrooms for research was carried out in the first half of July annually).

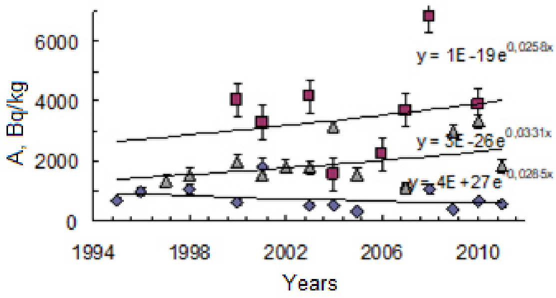

Fig. 4. Time trends in the ^{137}Cs content in mushrooms from ShNNP: *Boletus edulis* (◆), *Boletus badius* (■) and *Chanterelles* (▲).

As can be seen from the presented in Fig. 4 data, during 1994-2012, there is no single indication of a decrease in the content of ^{137}Cs in mushrooms from the ShNNP. A slight decrease of the specific activity of radiocaesium present in the fungal bodies of mushrooms over time is observed only for *Boletus edulis* (trend line $A(t) \sim e^{-0,0285t}$). In the case of *Chanterelle* and *Boletus badius*, there is even a tendency for its insignificant growth (corresponding trends: $A(t) \sim e^{0.0331t}$ for *Chanterelle* and $A(t) \sim e^{0,0258t}$ for *Boletus badius*). It should be noted that the statistical significance of these trends is low - the accuracy of the approximation of R^2 is 0.10 for Boletus edulis, 0.20 for *Chanterelle* and 0.04 for *Boletus badius* mushrooms.

The observed trends in the content of ^{137}Cs in mushrooms with CHNPP are correlated with the data, obtained by other authors for mushrooms from different regions of Europe. Thus, in [14] there is a decrease in the content of radionuclide in the fruit bodies of the *Rozitescape-ratus* mushrooms, selected in Denmark in 1986-1994; in the years to 2001, its content remained practically unchanged.

The study of mushrooms, selected in Sweden from 1986 to 1998, showed an increase in radiocaesium content over time; this gave rise to the author [15] to conclude that radioactive decay seems to be the only factor that will significantly affect on the ^{137}Cs content in mushrooms in the long term perspective.

About the actual increasing of the ^{137}Cs content in fruit bodies of mushrooms (including *Boletus edulis*), sampled in 2006-2001 on the territory of the Gomel region, Belarus, was reported in [16]. The authors notes a great influence of natural factors, first of all, weather-forming, on the degree of accumulation of radionuclides – in arid years with high temperatures its content in the fruit bodies of mushrooms increases.

Based on the results of studies on changes in the content of radioactive caesium in mushrooms selected in northern Italy, the authors [17] noted the practical absence of significant changes in the ability to accumulate radionuclide with time. Moreover, some kinds of mushrooms over time somewhat reduced the rate of accumulation of ^{137}Cs, while others, on the contrary, increased.

In [1] is noted that the depth of placement of mycelium in the soil influences the accumulation of radionuclide in mushrooms. For mushrooms whose mycelium are located in the surface layers of forest litter, in 1987-1994 there was a characteristic decrease in its content in the fruit bodies, whereas in mushrooms whose mycelium are located in the mineral layers – increase.

In addition, the authors [1] emphasize the need to consider that for the same kinds of mushrooms their mycelium can be located at different depths under different types of forest growth conditions, which leads to a change of the transfer factor T_f value up to 200 times for the same species of mushroom, depending on the type of soil.

According to [8], the contamination of the fruit bodies of mushrooms of the same species during the fruiting season significantly increases from 1,1 to 30 times in the autumn relative to the beginning of the season depending on the degree of soil contamination.

Conclusions

The results obtained by us, as well as the analysis of studies carried out by other researchers, confirm the fact that the accumulation of ^{137}Cs by mushrooms that growing in radioactive contaminated territories, other than the actual level of radioactive contamination of soils, affects their belonging to a certain species. Those are caused different depth of mycelium placement in the soil and the ability to absorb radionuclide from the soil. There is also a complex of natural factors, which significantly affect the ability of fungi to accumulate radionuclide from the nutrient substrate. Such factors are: types of soil types that determine the peculiarities of radionuclide contamination mushrooms and accessibility of ^{137}Cs to assimilation by mycelium; weather regimes of the territory and seasonal faults of their changes; the depth of placement of mycelium of mushroom rooms for the same species, which may also be different in soils of different types and under different weather conditions, etc. The complex effect of all these factors leads to a change (decrease or increase) of ^{137}Cs content in mushrooms over time, which is observed.

In predicting of changes in the state of radiocaesium contamination of mushrooms that grow on areas that have undergone radioactive contamination, it should be taken into account that the migration of radionuclide into the soil due to its consolidation on soil complexes plays a comparatively small role in reducing of the content of radiocaesium in its top near-surface layer. The results of studies that show the absence of an explicit tendency to reduce the contamination of fungi with radionuclide over time also should be taken into account. Given this, one can conclude that there is no weighty reason to hope that the ^{137}Cs contamination level for the vast majority of mushrooms which growing in the contaminated region will decreases in the near future markedly - obviously, this decreasing will be determined the natural decay of radionuclide mainly.

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