Acta Agrophysica, 2002, 67, 95-103

ACCUMULATION OF RADIONUCLIDES BY MUSHROOMS IN THE WESTERN REGION OF UKRAINE

V. Hrabovskyy, O. Dzendzelyuk

Department of Physics, Lviv Ivan Franko National University
Dragomanov 19 str., 79005 Lviv, Ukraine, e-mail: grabovsky@wups.lviv.ua

A b s t r a c t. Some results of a 5-year investigation of the radioactive contamination of mushrooms originating from the Western region of Ukraine are presented. Considerable variations of radiocaesium accumulation by mushrooms which belong both to different and the same species were noted. The corresponding radiocaesium transfer factors (a ratio of the caesium isotope content in mushroom and in soils at a given area) were calculated. For the mushrooms collected from the territory of the Shatsk National Natural Park (ShNNP, Volyn region) a sequence representing an increase of ^{137}Cs accumulation was complet sequence.

K e y w o r d s: mushrooms; radioactive contamination; radionuclides; radiocaesium

INTRODUCTION

The Chernobyl disaster, of April 26th, 1986 has been recognized as the greatest technogenic break-down in the history of mankind. It largely affected ecological condition of the environment [2,3]. During and directly after the break-down, radionuclides with a total activity of more than 50 MCi were thrown out into the environment from the emergency fourth block of the Chernobyl NP [3]. A resulting radioactive precipitations caused radioactive contamination of significant territories of our planet. Now, 16 years after the break-down isotopes of ^{137}Cs, ^{90}Sr, and also some representatives of a transuraniums, in particular plutonium and americium isotopes, are the most dangerous of them.

An analysis of the radioecological conditions of soils in Ukraine carried out for several years after the Chernobyl accident showed that its total territory is in fact more or less contaminated by the radioactive isotopes thrown out from the damaged block [3,11]. The most contaminated areas are the northern parts of Kyiv, Chernigiv, Zhytomyr, Rivne and Volyn regions. The ^{137}Cs contamination density in the soils of a significant part of these areas exceeds 37 kBq m^{-2}, reaching values of more than 1 MBq m^{-2} at places. The Western region of Ukraine, except for the polluted Rivne and Volyn regions, includes the Lviv region as the radiologically purest in Ukraine [11,12] and also relatively pure (except for some individual polluted territories) the Ivano-Frankivsk and Ternopil regions.

The Western region of Ukraine is characterised by a significant share of woods with specific flora and fauna in its territory. Soils of this region are characterized by high migration ability of radiocaesium and strontium deposited in them, hence a significant pollution of vegetation by the above radionuclides [7]. A special place among them is occupied by mushrooms, known as concentrators of microelements [7,13] (including radionuclides) from the environment vegetation. Consequently, they can be used as biological indicators of the radiological purity. In this article, some results of studies on the specific features of pollution with radiocaesium of edible mushrooms in the Western region of Ukraine are presented.

MATERIALS AND METHODS

Samples of edible mushrooms were investigated. They were collected in 1994-2000 in the territories contaminated, by the Chernobyl accident (Volyn, Rivne, Zhytomyr and Kyiv regions), and in the relatively pure territories (Lviv and Ivano-Frankivsk regions). It is known that the level of radionuclides accumulation in mushrooms depends to a large extent on the species [9,13] and sampling place and time [8]. Therefore, to eliminate the influence of temporary and territorial factors together with soil-climatic conditions and the influence of the sampling place, part of mushrooms studied was sampled each year in the first half of July from the same site in an area of 1 sq. km close the Pisochne Lake in the Shatsk Natural National Park (ShNNP) of the Volyn region. Soil samples for the determination of radionuclides specific activity and contamination density were selected there.

Preparation of the mushroom selected samples for analysis included separation of their stems and caps, then careful clearing off the remains rests of soil and vegetation, drying and homogenizing. Sample drying cleared external impurities. It was carried out in a drying cupboard at 80-90^{o}C for 24 h. Sample homogenization was carried out by the pulverization and careful mixing. Then, part of the sample necessary for the analysis was selected, weighed with an accuracy of 0.1 g, packed into measuring vials and placed in a spectrometer for the measuring.

Soil sampling at each of the sites selected was carrid out at five points (at the borders and in the centre of the spot selected) from a depth of 20 cm. A cylindrical soil sampler with a cylinder bore of 40 mm was used for sampling. Samples from each of the five points selected were cleared from organic and other impurities and then poured in one package. After drying, homogenization and weighing, a part with a volume of 1 l was separated for research from the mass of soil, and then weighed, placed in Marinelli beaker and located in the measuring chamber of the spectrometer.

The gamma-emitted radionuclide content in the samples was analyzed by a gamma-spectrometer using a Ge(Li)-detector with resolution (FWHM) of 2.5 keV at 1332 keV. For the reduction of the influence of external background radiation, the detector was surrounded by a leaden protection with a thickness of 50 mm. Measuring times were set to ensure an error of not more than 10% for the special activity determination at the geometry of the measuring array used.

RESULTS AND DISCUSSION

The results of the ^{137}Cs content determination in stems and caps of edible mushrooms selected in the vicinity of the Pisochne Lake (ShNNP) in July 2000 are given in Table 1. Except for ^{137}Cs, only ^{134}Cs and ^{40}K isotopes were identified in all mushrooms investigated, though there are some other gamma-emited isotopes in the soil, members of the uranium and thorium natural radioactive series. The special activity of ^{134}Cs found in the most contaminated samples (*Boletus subtomentosus* L. ex. Fr. and *Tulopilus felleus* (Fr.) Karst.) is 180 times higher than the ^{137}Cs content in them. This coincides with the ratio expected (about 183) of these radionuclides activities in the environment after the Chernobyl fallout during the time of measurements. It proved that their origin lies in the Chernobyl failure.

In order to estimate mushroom contamination by radiocaesium, an accumulation coefficient (sometimes referred to as a concentration factor) K_a and the transfer coefficient K_t (called an aggregation factor) [7,10] were used. They were calculated according to the formula:

$$K_a = \frac{A_p}{A_s}, \qquad K_t = \frac{A_p}{S_s}, \tag{1}$$

where A_p, A_s are specific activities of radionuclides in plant and soil (in Bq kg^{-1}), respectively, and S_s contamination density of a soil (in kBq m^{-2}). For the calculations of the above parameters, we determined average values of the specific activity

^{137}Cs (A_s = 19.7 Bq kg^{-1}) and density of pollution (S_s = 4.85 kBq m^{-2}) for the site where the mushrooms investigated were sampled.

The results presented in Table 1 are grouped for two kinds of mushrooms - lamellar and tubular – according to the increase of their ability to accumulate radiocaesium from the soil. Accumulation ability in the mushrooms examined increase according to the following sequence: *Macrolepiota procera* (Fr.) Sing. – *Boletus edulis* Fr. – *Boletus aurantiacus* Fr. – *Cantharellus cibarius* Fr. – *Boletus luteus* L. ex. Fr. – *Boletus Scaber* Fr. – *Boletus badius* Fr. – *Boletus subtomentosus* L. ex. Fr. – *Tulopilus felleus* (Fr.) Karst. Beside a species-specific dependence of the accumulation level for radiocaesium, significant differences in the accumulation by mushrooms of the same family were observed. It means that, among the representatives of the *Russula* family, radiocaesium accumulation coefficient increased in the following sequence: *Russula badia* Quel. *Russula xerampelina* (secr.) Fr. – *Russula heterophylla* Fr. – *Russula paludosa* Britz. – *Russula maculata* Quel. – *Russula claroflava* Grove. – *Russula farinipes* Rom arud Britz., increasing its value by more than twice.

Table 1. Special activity *(A)*, accumulation (K_a) and transfer (K_t) coefficients of ^{137}Cs in caps and stems of some species of mushrooms from ShNNP

Species of mushrooms	A, Bq kg^{-1}			K_a; K_t			
	A_c (caps)	A_{st} (stems)	A_c/A_{st}	K_c (caps)	K_t (caps)	K_c (stems)	K_t (stems)
	Tubular mushrooms						
Boletus edulis Fr.	618	364	1.70	31.4	127.4	18.5	75.0
Boletus aurantiacus Fr.	706	432	1.63	35.8	145.6	21.9	89.1
Boletus luteus L. ex. Fr.	2190	1060	2.06	111.2	451.5	53.8	218.6
Boletus Scaber Fr.	2830	1370	2.05	143.6	583.0	69.5	282.5
Boletus badius Fr.	4040	2310	1.75	205.1	833.0	117.3	476.3
Boletus subtomentosus L. ex. Fr.	7480	3510	2.13	379.7	1542.3	178.2	723.7
Tulopilus felleus (Fr.) Karst.	10100	7840	1.29	512.7	2082.5	398.0	1616.5
	Lamellar mushrooms						
Cantharellus cibarius Fr.	1980	1710	1.16	100.5	408.2	86.8	352.6
Macrolepiota procera (Fr.) Sing.	49	27	1.81	2.5	10.1	1.4	5.6
Russula badia Quel.	1990	1490	1.34	101.0	410.3	75.6	307.2
Russula xerampelina (secr.) Fr.	2480	1960	1.27	125.9	511.3	99.5	404.1
Russula heterophylla Fr.	4580	1550	2.95	232.5	944.3	78.7	319.6
Russula paludosa Britz.	5110	1960	2.61	259.4	1053.6	99.5	404.1
Russula maculata Quel.	5170	1910	2.99	262.4	1066.0	96.9	393.1
Russula claroflava Grove.	5790	1860	3.11	293.9	1193.8	94.4	383.5
Russula farinipes Rom arud Britz.	8310	3350	2.48	421.8	1713.4	170.1	690.7

A comparison of the results obtained with those known from literature, showed a good agreement in our species-specific dependence of radiocaesium accumulation ability data with results for the mushrooms from Poland [9]. However, they differ slightly from the results given for mushrooms from the Ukrainian Polesye [7,13]. Probably, these discrepancies are caused by different conditions of mushrooms growing. It is known [8] that the ability of mushrooms to accumulate radionuclides depends on the collection place and time.

For all the species of mushrooms examined, the content of the radiocaesium accumulated in caps exceed the content in stems without any exceptions. This is in agreement with results given in [4]. The value of such exceeding is within limits of 1.3-2.1 times for tubular mushrooms and 1.2-3 times for lamellar mushrooms. Obviously, such a situation may be caused by different functional assignments of these parts of mushrooms and, accordingly, by distinctions in their biological and chemical structures. An indirect confirmation of this may be the fact that ^{40}K content in the mushrooms caps, exceeded the content in their stems.

The results obtained show that even in the territories with a low contamination level (contamination density for the radiocaesium in the soils of the Park territory did not exceed 12 kBq m^{-2}), the factors of radiocaesium accumulation in mushrooms of various species varied in a range from 10 (for *Macrolepiota procera* (Fr.) Sing.) to up to more than 2000 (for *Tulopilus felleus* (Fr.) Karst.). Accordingly, contents of ^{137}Cs in the mushrooms investigated ranged from 30-50 Bq/kg up to more than 10000 Bq kg^{-1} (in terms of dry weight). Taking into account the allowable levels of radiocaesium contamination of food currently valid in Ukraine, on the allowable limits for dry mushrooms (specific activity not higher than 2500 Bq kg^{-1}), it can be seen that not all the mushrooms studied are permised for the use in the food-stuff. Besides, the results of radiocaesium activity measuring in mushrooms collected in 1995-2000 showed that were no appreciable essential temporary changes in the radiocaesium content. *Boletus edulis* mushrooms sampled in the territory of the Park during one and the same period of time each year (in the first half of July) showed the following values of the specific activity for ^{137}Cs: 1995: 684-847; 1996: 769-984; 1997: 779-876; 1998: 655-1030; 2000: 669-1030 Bq kg^{-1}. Slightly lower values of the radiocaesium specific activity in the mushrooms selected in 1995-1997 can be caused by a drier climate – spring and summer in these years, especially in 1995, were dry. As marked in [7], dry climate reduces migration of radionuclides from the soil to the fruit body of mushrooms.

Such a situation is caused by the fact that total radiocaesium pollution of the forest litter and its superficial 10-cm layer of soil changed only slightly in these years. Both data known from literature and our own data for the vertical radiocaesium migration in the soils of the Park, confirmed the above. For example, in paper [6], the authors stated that on the basis of their 15-year supervision they were able to prove that ^{137}Cs migrates from the forest litter to the soil. Then it is fixed in the top five-centimetre of the soil layer. Further appreciable migration in the soil layers below is not marked. Therefore, one can hardly hope that radiocaesium contamination of plants from the Park, including mushrooms, would be significantly reduced at the expense of their migration into the deeper soil layers at least, not in the nearest couple of years. Most probably, such a reduction can be only caused by a process of radiocaesium content decrease in the top layers of the soils due to its natural decay.

Some results of measurements of the gamma-emitted radionuclide content in *Boletus edulis* collected at various sites of the Western region of Ukraine and some regions of Kyiv and Zhytomyr areas during 1995-2000 are shown in Table 2. Contamination density levels of the soils from the territories where the mushrooms investigated grew before sampling are also presented. Data concerning contamination density of soils are partially taken from earlier papers [5,11,12]. Data on contamination density levels of the soils from the Lviv area, the ShNNP and Bystrychi village (the Berezne district of the Rivne area) were obtained by the present authors.

On the whole, the values of mushromms contamination by ^{137}Cs correlated well with the contamination data for the corresponding soils. The latter confirms the conclusion that the results for the radioactive contamination determined for mushrooms may be used for a comparative analysis of levels of radioactive contamination of soils. However, for a more correct estimation of the level of environment contamination established on the basis of content of radioactive elements in mushrooms one has to take into account the following factors:

- for a comparative analysis of the pollution level of soils, it is necessary to use mushrooms of the same species, as various species, even if collected in the same territory, can concentrate radiocaesium in a different way;
- mushrooms absorb radiocaesium for soils in the soluble form; therefore the level of their contamination gives mainly information on the dissolved phase of ^{137}Cs in the forest litter and in the superficial layer of the soil (with depth up to 10 cm) [6];
- due to migration processes of radionuclides in the soils, ^{137}Cs can move down to the lower horizons and not be accumulated in the mushroom fruit bodies for which the depth of mycelium does not usually exceed 10 cm.

Table 2. Contents of radioactive caesium and potassium in *Boletus edulis*, selected in 1995-2000 in the Western region of Ukraine (at Bq kg^{-1} of dry weight)

Sites of selection	Year	Activity, Bq/kg			S_s, ^{137}Cs
		^{134}Cs	^{137}Cs	^{40}K	kBq m^{-2}
Shklo, Javoriv distr., Lviv region	1996	2	93	889	1-2
Wowche, Turka distr., Lviv region	1997	-	93	1080	2-3
Wowche, Turka distr., Lviv region	1998	-	45	1020	2-3
Skole, Lviv region	1998	-	119	938	3-4
Potelych, Zhovkva distr., Lviv region	1998	-	218	971	2-3
Kuratova, Kosiv distr. Iv-Frankivsk region	1997	-	140	886	4-10
Sheshory, Kolomyja distr., Iv-Frankivsk region	1997	-	554	1160	4-10
Lake Svityaz', ShNNP, Volyn region	1997	-	779	911	5-12
lake Pisochne, ShNNP, Volyn region	1995	13	684	1000	5-12
lake Pisochne, ShNNP, Volyn region	1996	10	769	912	5-12
Lake Pisochne, ShNNP, Volyn region	1998	10	939	847	5-12
Swalovychi, Ljubeshiv distr., Volyn region	1998	72	7860	1050	100-185
Manewychi, Volyn region	1999	35	4980	865	100-185
Mostyshche, Kamin'-Kashyrskyy distr., Volyn region	2000	-	1040	741	10-20
Mala Sowpa, Korets distr., Rivne region	1997	-	843	916	<18
Wesnjane, Korets distr., Rivne region	2000	-	1650	849	10-20
Bystrychi, Berezne distr., Rivne region	1997	154	11800	912	30-44
Udrytsk, Dubrowytsja distr., Rivne region	1997	520	83300	901	185-550
Dubrowa, Malyn distr., Zhytomyr region	1986	20	2990	680	10-20
Baraniwka, Zhytomyr region	1997	-	593	645	4-10
Stary Sokoly, Iwankiw distr., Kyiv region	1999	52	7490	1060	

Besides, it is necessary to take into account that different parts of the mushroom fruit body accumulate radionuclides differently: the level of accumulation in caps is usually considerably higher than in stems. That is why one should use data for identical parts of the mushroom fruit bodies for the analysis of the contamination level. Moreover, because of different abilities to accumulate radionuclides, caps should be advantageous.

Results on the content of ^{90}Sr in one of the most contaminated samples of the mushrooms showed that the corresponding specific activity did not exceed one percent of the value characteristic for the specific radiocaesium activity. The measurements were also carried out with a scintillation beta-spectrometer according to recognised techniques [1] used at the Radiological Department, Lviv Regional Sanitary-Epidemiological Station. The results agreed well with data of paper [11]. They allow to draw a conclusion that only a gamma-spectral analysis can be used when determining radiological cleanliness of mushrooms in the territory of the Ukrainian Polesye after pollution originating from the Chernobyl accident. It would allow to get a quite accurate representative estimation of the radiological cleanliness of mushrooms without the carrying out of more laborious and expensive beta-spectral analyse.

CONCLUSIONS

The results of gamma-spectrometric measurements of the radionuclide accumulation in edible mushroom originating from the territory of Western Ukraine after the Chernobyl disaster, showed that:

1) ^{137}Cs specific activities in the samples investigated changed from 100 Bq kg^{-1} to more than 10000 Bq kg^{-1} (in dry weight).

2) It was also found that the radiocaesium accumulation factor for caps of all mushroom samples tested, exceeded those for stems by 1.4 to 2.0 times.

3) On the whole, the values determined for ^{137}Cs contamination rates in the mushrooms are well coordinated with the contamination data for the soil of the same location.

4) Beta-spectrometric analysis of some highly contaminated samples showed that ^{90}Sr content did not exceed 1% found for ^{137}Cs in any of the mushroom samples investigated. The above is a possibility to get an actual estimation of the mushroom samples radiological cleanliness (together with corresponding soils, too), without performing a more laborious and expensive beta-spectrometric analysis.

ACKNOWLEDGEMENTS

The authors are grateful to the engineer H. Ducjak of the Nonlinear Optics Chair, Lviv Ivan Franko National University for her assistance in gamma-spectral measurements, as well as to the manager I. Koljanchikov, of the Radiological Department, Lviv Regional Sanitary-Epidemiological Station, and the engineer O. Pidsadnyuk for their support and assistance during the beta-analysis of samples.

REFERENCES

1. Activity of beta-radiating radionuclides in accounting samples. A technique of measurements fulfilment with using of scintillation spectrometers and software AK1 (in Russian). Kiev, 1998.
2. **AnspaughL.R., Caltin R.J., Goldman M.:** The global impact of the Chernobyl reactor accident. Science, 242, 1988.
3. Chernobyl disaster (in Russian). (Ed. Baryakhtar V.G.). Naukova dumka, Kiev, 1995.
4. **Dietl G.:** Zur Verteilung radioactiver Casiumnuclide im Pilzfruchtkörper. Z. Mycol., 55, 1, 1989.

5. **Korotun I.M., Korotun L.I.**: Geography of Rivne region (in Ukrainian). Rivne, 1996.
6. **Kozhevnikova T.L., Mishenkov N.N., Martyushova L.N., Krivolutskiy D.A.**: Accumulation of ^{90}Sr and ^{137}Cs by fruit bodies of mushrooms (in Russian). Ekologiya, 6, 1993.
7. **Krasnov V.P.**: Radioecology of the forests of Ukrainian Polissya (in Ukrainian). Zhytomyr, 1998.
8. **Meijer R.J., Aldenkamp F.J., Jansen A.E.**: Resorption of caesium radionuclides by various fungi. Oecologia, 77, 2, 1988.
9. **Mietelski J.W.** *et al.*: Radioactive contamination of Polish mushrooms. Sci. Total Environ., 157, 1994.
10. **Mietelski J.W., Jasińska M.**: Radiocesium in bilberries from Poland: comparison with data for mushroom samples. Journal of Radioecology, 14, 1996.
11. Protection of the environment in Ukraine. 1994-1995 (in Ukrainian). Ed. Rayevskogo, Kyiv, 1997.
12. **Vlokh O.G., Grabovsky V.A., Dzendzeljuk O.S.**: The investigation of degree of soil contamination by Cs-137 of populated areas in Lviv region (in Ukrainian). Ekotekhnologiyi i resursosbyeryezheniye, Kiev, 4, 1994.
13. **Wasser C.P., Grodzinska G.A., Lyugin V.O.**: Accumulation of radioactive elements by macromycetes of Ukrainian Polissya (in Ukrainian). Ukr. Bot. Z., 49, 5, 1992.

AKUMULACJA RADIONUKLIDÓW PRZEZ GRZYBY JADALNE W REGIONIE ZACHODNIEJ UKRAINY

V. Hrabovsky, O. Dzendzelyuk

Katedra Fizyki Lwowskiego Uniwersytetu Narodowego im. Iwana Franki
ul. Dragomanov 19, 79005 Lwów, Ukraina, e-mail: grabovsky@wups.lviv.ua

S t r e s z c z e n i e. Pięcioletnie badania zawartości radionuklidów w grzybach pochodzących z regionu Zachodniej Ukrainy (Szacki Park Narodowy) wykazały ich radioaktywność ^{137}Cs w granicach 100-10.000 Bq kg^{-1} w s.m. Akumulacja radiocezu była przy tym 1,4 do 2,0 razy większa w kapeluszach niż w trzonach grzybów i w całości była dobrze skorelowana z zawartością ^{137}Cs w glebie.
S ł o w a k l u c z o w e: grzyby, zanieczyszczenia radioaktywne, ^{137}Cs, Zachodnia Ukraina