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Distribution of ^{137}Cs in soils and wild mushrooms from Bulgaria

Radoslava Lazarova[*] and Milena Hristozova

Agricultural Academy, „ N. Poushkarov" Institute of Soil Science, Agrotechnologies and Plant Protection, Testing Laboratory of Radioecology and Radioisotopes Research, 1331 Sofia, Bulgaria
[*]*Corresponding author:* lazarova.radi@gmail.com

Abstract

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The content of technogenic ^{137}Cs was studied in soils and wild mushrooms from Bulgaria.

The specific activity of the radionuclide varied between 2.0±0.5 and 35±3 Bq/kg in the soils from Northern Bulgaria and between 4±1 and 270±10 Bq/kg in the soils from southern Bulgaria. The significant standard deviation calculated indicated firm heterogeneity in pollution, particularly in the southern mountainous part of the country.

The highest accumulation of Cs-137 was found in wild mushrooms from the species *Hydnum repandum* – 890±90 ÷ 1500±50 Bq/kg, *Hydnum imbricatum* – 1250±60 ÷ 2860±150 Bq/kg, and *Cantharellus cinereus* – 890±90 ÷ 2030±100 Bq/kg. Lower values were measured in *Boletus edulis* – < 1÷55±5 Bq/kg (fresh weight) and 10±2 ÷ 400±20 Bq/kg (dry weight), and *Cantharellus cibarius* <1 to 55±6 (fresh weight) and 15± to 190±20 Bq/kg dry weight. The lowest levels of radiocaesium were recorded in *Marasmius oreades* – <1 to 10±2 Bq/kg, *Morchella esculenta* – <1 to 20±3 Bq/kg, and *Craterellus cornucopioides* – <1 to 20±4 Bq/kg.

The annual effective dose from consuming mushrooms with ^{137}Cs content exceeding the EU limit of 600 Bq/kg was calculated. It was found that its contribution to the annual effective dose from all radioactive sources is insignificant, and it is not hazardous to the population from a radiological point of view. However, their regular consumption is not recommended.

Keywords: ^{137}Cs; soil pollution; radioactivity of wild mushrooms

Introduction

Research on the contamination of soils in Bulgaria with artificial radioactive elements, mainly ^{90}Sr and ^{137}Cs, began in 1960. Until the Chernobyl accident, the main source of radioactive pollution of the soils in the country was nuclear weapon tests carried out in the atmosphere and the oceans. The accident changed radically the radioactive status of the soils in Bulgaria (Naydenov and Staneva, 1987). Since then, regular research on undisturbed soils from a set monitoring network has been carried out by the Laboratory of Radioecology and Radioisotopes Research at the „N. Poushkarov" Institute. It shows that thirty years later, one half-life of ^{137}Cs, the radionuclide, is still being detected in the soils of Bulgaria (Yordanova et al., 2014; 2016), representing a potential risk for plant contamination and a radiation hazard to humans along the food chain.

Wild mushrooms are well known for their ability to accumulate radionuclides and are often used as indicators of radioactive contamination (Gillett and Crout, 2000; Tucaković et al., 2018). They are also highly valued for their taste qualities, and their consumption may pose a radiological risk to the population.

The study aims to present the results on the content of cesium-137 in soils from the monitoring network in Bulgaria over recent years, and to investigate radionuclide accumulation in different species of wild mushrooms. Additionally, it seeks to assess the risk from an additional dose load to the population from the consumption of radioactive mushrooms.

Materials and Methods

The regular radiological monitoring of soils in Bulgaria is conducted according to a sampling strategy described elsewhere (Yordanova et al., 2014). Undisturbed soils from 35 sampling points, including the 30 km area around Kozloduy NPP and along the Danube River, are collected from the monitoring network in North Bulgaria. Undisturbed soils from the Rhodopes and along the Mesta River valley were sampled at 34 points in Southern Bulgaria. Soil samples are homogenized, dried at 80°C, and sieved through a 2 mm sieve.

The number of wild mushroom samples from different regions of the country analyzed in the period 2016-2023 is 206. The mushrooms are from the species *Boletus edulis*, *Cantharellus cibarius*, *Cantharellus cinereus*, *Hydnum repandum*, *Hydnum imbricatum*, *Marasmius oreades*, *Morchella esculenta,* and *Craterellus cornucopioides*.

^{137}Cs in soils and mushrooms is determined gamma-spectrometrically by the full energy peak at 661.6 keV. A DSA 1000 multichannel analyzer (produced by CANBERRA) coupled to a germanium detector with 30% efficiency and 1.8 keV resolution at the Co-60 full-energy peak (1332.2 keV) is used. Samples are measured for 60 000 s in 450 mL Marinelli beakers. The spectrum is analyzed using GENIE-2000 software. The expanded uncertainty (U) is calculated in Bq/kg at a 95% confidence interval (k = 2). Certified reference materials of gamma-emitting radionuclides are used for energy and efficiency calibration.

Results and Discussion

Before the Chernobyl accident in 1986, the average value of ^{137}Cs activity in soils of Northern Bulgaria (around the Kozloduy Nuclear Power Plant and along the Danube River) was 10 Bq/kg, and in southern Bulgaria (the Rhodopes) – 26 Bq/kg (Naydenov, 1986). After the accident, the amount of radiocaesium deposition increased significantly.

The regular studies of undisturbed soils included in the monitoring network of Bulgaria carried out by the Laboratory after the accident in the period 1986 – 2015, found the average values of the radionuclide varied between 40 and 60 Bq/kg for North Bulgaria and were 2 to 5 times higher in the mountainous regions of South Bulgaria – 160 and 280 Bq/kg respectively, with standard deviation between 30 and 60% showing the significant inhomogeneity of the deposition (Yordanova, et al., 2016). Although the activity of ^{137}Cs in the surface soil layer is decreasing, primarily due to natural decay, data collection on the activity and dynamics of the radionuclide continues.

The analyses of Cs-137 content in soils carried out between 2015 and 2022 are presented in Figure 1 for North Bulgaria and Figure 2 for South Bulgaria, respectively. The value is calculated by averaging the results obtained from soil samples collected from the points of the monitoring network in North and South Bulgaria, respectively.

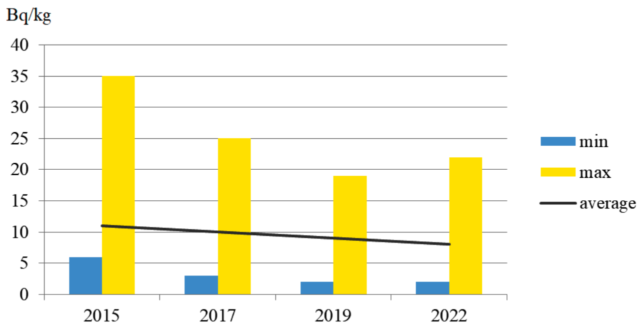

Fig. 1. ^{137}Cs content in the soils of North Bulgaria

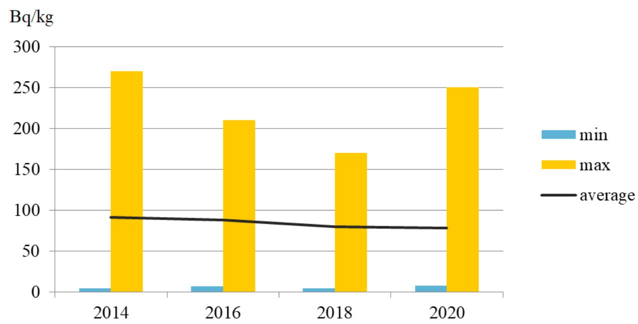

Fig. 2. ^{137}Cs content in the soils of South Bulgaria

The results obtained show that ^{137}Cs activity in the soils of North Bulgaria reaches the levels registered before the Chernobyl accident. However, it remains several times higher in South Bulgaria, particularly in the Rhodopes. This could probably be explained by the difference in altitude, which determines the amount of precipitation, as well as the fact that the radioactive cloud passed over the southern territory of the country twice in 1986.

The standard deviation of the values calculated for each year varies between 27% and 70% for the soils from North Bulgaria and between 72% and 88% for the soils from South Bulgaria, showing a significant variation in ^{137}Cs activity. The strong inhomogeneity of pollution, which is still found even within small areas, is probably due to the relatively short time of deposition in combination with heavy rains that occurred in May 1986 after the Chernobyl accident (Yordanova et al., 2007a).

The habitat of wild mushrooms is usually found in forest areas in mountainous regions. The mycelium is located mainly in the upper layers of the soil. The forest floor can retain a significant portion of radiocaesium, as its content increases exponentially with the increase in forest floor thickness (Zhiyanski et al., 2008). The principal amount of the radionuclide is found in the surface soil layer, 0-5 cm deep (Zhiyanski and Sokolovska, 2006).

Furthermore, mushrooms growing in sandy, mountainous soils with coniferous vegetation are reported to contain higher ^{137}Cs concentrations compared to those from deciduous and meadow vegetation areas (Baeza and Guillén, 2007). This could be explained by the smaller amount of clay fraction in sandy soils and the lower pH of coniferous forest soils, due to the presence of a greater amount of fulvic acids, which facilitates the bioavailability of radiocaesium compared to deciduous and meadow soils, characterized by a more clayey mechanical composition and higher pH.

The type of mushrooms and their feeding methods (saprophytic, mycorrhizal, and parasitic) are also essential. It is reported that mycorrhizal fungi accumulate radiocesium to a greater extent than saprophytic fungi (Duff and Ramsey, 2008). Therefore, it was of interest to analyze the distribution of ^{137}Cs in various mushroom species.

The percentage distribution of various mushroom species analyzed in the study is presented in Figure 3.

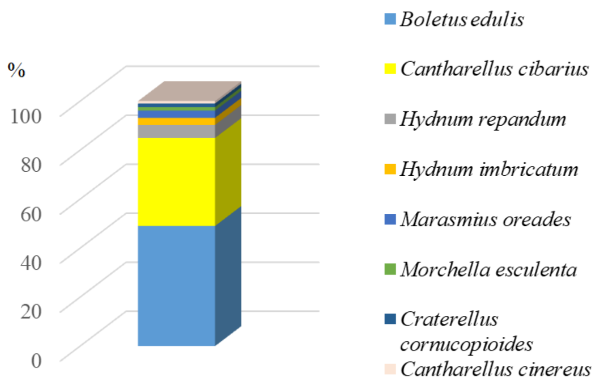

Fig. 3. Percentage distribution of tested wild mushrooms

According to EC Regulation 2020/1158 (EC Implementing Regulation 2020/1158, 2020), the specific activity of ^{137}Cs in mushrooms shall not exceed 600 Bq/kg.

In 8% of the samples, the radionuclide content was below the minimum detectable activity of 1 Bq/kg. In the rest of the samples, well-detectable amounts were registered, and in 5% of all tested mushrooms, specific activity above the norm of 600 Bq/kg was measured.

Accumulation of radioactive elements in plants depends on the type of radionuclide, its amount in the soil, the soil characteristics, and the type of plant itself.

Regarding the type of radionuclide, Cs-137 is part of the alkaline earth elements group, with chemical properties similar to those of potassium. This group member is one of the main inorganic macroelements necessary for plant growth. In addition, cesium compounds are characterized by good solubility in water, which facilitates their transfer from the soil solution to plants.

In the present study, the lowest activity concentrations of caesium-137 were found in the mushrooms of the species *Marasmius oreades* (<1 ÷10±2 Bq/kg), *Morchella esculenta* (<1 ÷20±3 Bq/kg), and *Craterellus cornucopioides* (<1 ÷20±4 Bq/kg).

Relatively low levels of radiocaesium were recorded in *Cantharellus cibarius* (fresh and dried) – Figure 4. The values ranged from <1 to 55 ± 6 Bq/kg in the fresh samples and from 15 ± 3 to 190 ± 20 Bq/kg (median: 70 Bq/kg) in the dried samples.

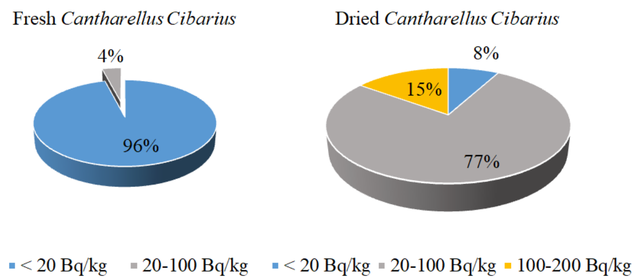

Fig. 4. Content of ^{137}Cs in fresh and dried *Cantharellus cibarius*

Buletus species are highly valued for their taste. *Buletus edulis* is one of the most common wild mushrooms collected in Bulgaria and is often exported to other European countries. It primarily grows in deciduous forests, forming a network of hyphae that surrounds the tree roots in a so-called ectomycorrhizal symbiosis. The *Buletus* species are characterized by a medium to high ability to accumulate radiocaesium, varying significantly from one species to another (Duff and Ramsey, 2008). In the present study, the content of cesium-137 measured in *Buletus edulis* was between <1 and 55 ± 5 Bq/kg in fresh mushrooms and between 10 ± 2 and 400 ± 20 Bq/kg, with a median of 60 Bq/kg, in dry mushrooms (Figure 5).

For comparison the distribution of radiocaesium in mushrooms of the same specie studied in the Laboratory in 2005 (19 years after the Chernobyl accident) was as follows: fresh

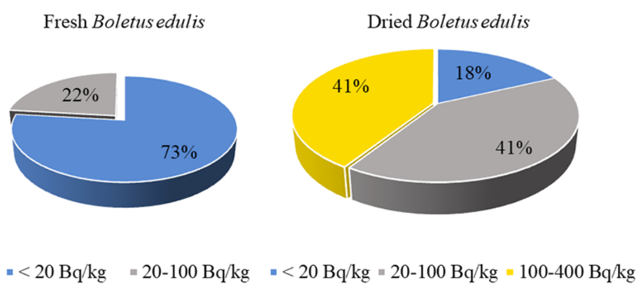

Fig. 5. Content of ^{137}Cs in fresh and dry *Boletus edulis*

samples: < 20 Bq/kg – 65%; 20 ÷ 100 Bq/kg – 24% and 100 ÷ 600 Bq/kg – 11%; dry samples: <20 Bq/kg -12%, 20÷100 Bq/kg – 34%, 100 ÷ 600 Bq/kg – 51%. In 3% of the samples, the cesium-137 content exceeded 600 Bq/kg (Yordanova et al., 2007b).

It can be seen that, in the present study, 30-37 years after the Chernobyl accident, when more than half of the cesium-137's half-life has passed, the specific activity of the radionuclide detected in fresh mushrooms was below 100 Bq/kg, and in dried mushrooms, below 400 Bq/kg. Activity concentrations over the permissible level of 600 Bq/kg were not detected in *Buletus edulis*.

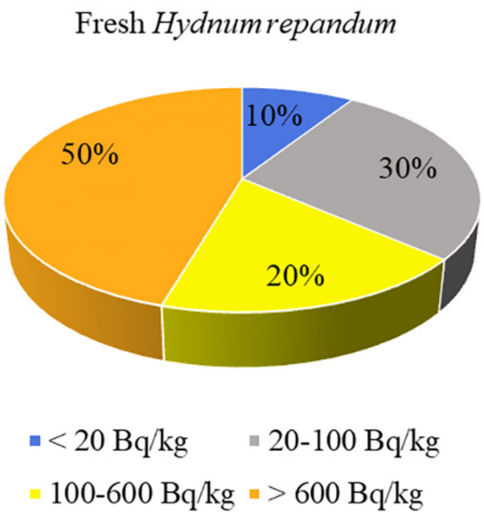

Fig. 6. Content of ^{137}Cs in fresh *Hydnum Repandum*

However, higher ^{137}Cs accumulation was found in *Hydnum repandum* sp., characterized by mycorrhizal feeding as well. The specific activity of the radionuclide varied between 15±2 and 1500±50 Bq/kg (median 700 Bq/kg) – Figure 6. It can be seen that the radiocaesium content measured in half of the samples exceeded the maximum permissible level of 600 Bq/kg.

High accumulation of cesium-137 was also detected in the species *Hydnum Ibricatum* and *Cantharellus cinereus*; the specific activity in the former ranged from 110±15 to 2860±150 Bq/kg, and in the latter, two samples were measured with activity concentrations of 890±90 and 2030±100 Bq/kg, respectively.

The contribution from the consumption of mushrooms with a content of ^{137}Cs over 600 Bq/kg to the annual effective dose was calculated to assess the risk of additional dose load to the population according to the following formula (Kalač, 2001):

$$E = Y \times Z \times dk,$$

where Y – consumption of mushrooms for one year (in kg per person);

Z – the specific activity of the radionuclide (Bq/kg);

Dk – dose factor whose value for ^{137}Cs is 1.3×10^{-8} Sv/Bq (ICRP, 2012).

The values were calculated based on an average annual consumption of 0.3 kg of mushrooms by the Bulgarian population (Bulgarian Food Safety Agency, 2011) (Table 1).

The world average annual effective dose to the population from all natural radioactive sources is approximately 2.4 mSv. As can be seen from the results obtained, the contribution from the consumption of mushrooms with ^{137}Cs content exceeding 600 Bq/kg varies between 0.15% and 0.47%, and can be considered insignificant. However, their regular consumption in greater quantities is not recommended.

Table 1. Effective dose per year (E) from the consumption of mushrooms with specific activity of radiocaesium over 600 Bq/kg

Fresh *Hydnum Repandum*		Dry *Hydnum Ibricatum*		Dry *Cantharellus Cinereus*	
Specific activity	E	Specific activity	E	Specific activity	E
Bq/kg	mSv/year	Bq/kg	mSv/year	Bq/kg	mSv/year
1050	0.004	2860	0.011	2030	0.008
1500	0.006	2600	0.010	890	0.003
890	0.003	1250	0.005		
1250	0.005				
920	0.004				

Conclusion

The present research on the content of radiocesium in soils from the monitoring network in Bulgaria reveals that the radionuclide activity reaches values comparable to those measured before the Chernobyl accident in Northern Bulgaria. However, they remain several times higher in South Bulgaria and the Rhodopes in particular, with high standard deviation from the average, showing the heterogeneity of contamination.

The activity of the radionuclide does not exceed the maximum permissible norm of 600 Bq/kg in the majority of the analyzed wild mushrooms collected from the country. Accumulation of Cs-137 is detected in the species *Hydnum Repandum*, *Hydnum Ibricatum,* and *Cantharellus Cinereus*. This is probably due to both the specific characteristics of the mushrooms and the areas with higher radioisotope content from which they were collected, such as high mountain ranges with coniferous forests. The results obtained indicate that the contribution of consuming these mushrooms to the annual effective dose is insignificant, and they do not pose a radiological risk to the population. However, their consumption in excess quantities is not recommended.

The regular monitoring of soil and wild mushroom radioactivity is also recommended, as the radiocesium content in Bulgarian soils remains high, especially in mountainous areas, and its activity distribution is inhomogeneous.

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