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THE ACCUMULATION OF HEAVY METALS BY MACROMYCETES IN BREST REGION OF THE REPUBLIC OF BELARUS

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A research team has analysed heavy metals accumulation by various species of wild edible macromycetes, which grow within the boundaries of contaminated and conditionally background territories of natural forest ecosystems of Brest region of the Republic of Belarus. It has been found that within the boundaries of the studied territory, the concentration of heavy metals in fruit bodies of macromycetes is more species-specific and environment dependent than the nature of the substrate and the place of growth. Mycological products, collected within both technogenous polluted and conditionally background territories of Brest region are contaminated with heavy metals, which form the following ranked series in terms of the content of macromycetes in fruit bodies: Zn > Cu > Cd > Pb > Ni > Co. Consumption of mycological products with the following levels of contamination: Cd – 8.4–16.2 MAC; Zn – 6.5–11.0 MAC; Cu – 2.5–5.2 MAC; Pb – 8.4–16.2 MAC; Ni and Co – 1.5 MAC throughout life can lead to the increase in carcinogenic and non-carcinogenic health risks. The level of individual carcinogenic risk from consuming macromycetes contaminated with lead and cadmium is $0.1–3.4 \times 10^{-2}$. Such a risk according to the international criterion scale is assessed as high, not acceptable for production conditions and for the population, and requires measures to be taken for its elimination or reduction.

Keywords: carcinogenic and non-carcinogenic risk, contamination, forest, heavy metals, macromycetes, soil

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INTRODUCTION

In the Republic of Belarus, mushroom produce is traditionally consumed by a large part of the population mostly due to its easy accessibility and low prices. Favourable natural conditions and a high level of afforestation stipulate the fact that the average annual mushroom biological and exploration reserves amount to 59.2 thousand tons and operational to 26.6 thousand tons respectively (Baginskiy, 2007; Koshedub, 2017). In addition, edible macromycetes prove to be a balanced natural complex of biologically active substances, essential amino acids and unsaturated omega-3 fatty acids. They also contain more than 20 macro- and microelements and A, B, E, D, PP vitamins (Yashchenko, 2012; Myslyva, Bilyavskiy, 2016).

However, under the unfavourable ecological conditions, due to the ever-growing anthropogenic impact on the environment, the mushroom produce can be heavily contaminated and damaging for the health of the population, since the mycobiota has a strong capacity for accumulating pollutants, and heavy metals in particular. Even 31 years after the Chernobyl disaster, the contamination of wild mushrooms with Cs^{137} in Belarus remains at a high level. According to the State Agency for the Protection and Monitoring of Forests "Bellesozashchita" the average content of this radionuclide in the fruit bodies of mushrooms growing in Brest region exceeds 1600 Bq/kg.

A review of available literature revealed that in Belarus there are practically no studies based on a long-term systematic research of the heavy metals accumulation in mushrooms and hazard assessment for humans consuming contaminated mycological products. However, the determination of heavy metal concentration in the fruiting bodies of mushrooms is essential in dietary intake studies: this acute problem is widely highlighted in scientific literature in many other countries (Wasser et al., 2012).

The purpose of the research was to study the peculiarities of Zn, Cu, Mn, Pb, Cd, Ni and Co accumulation in certain species of wild edible macromycetes with a tubular and lamellar hymenophore, which grow within the boundaries of contaminated and conditionally background territories of natural forest ecosystems

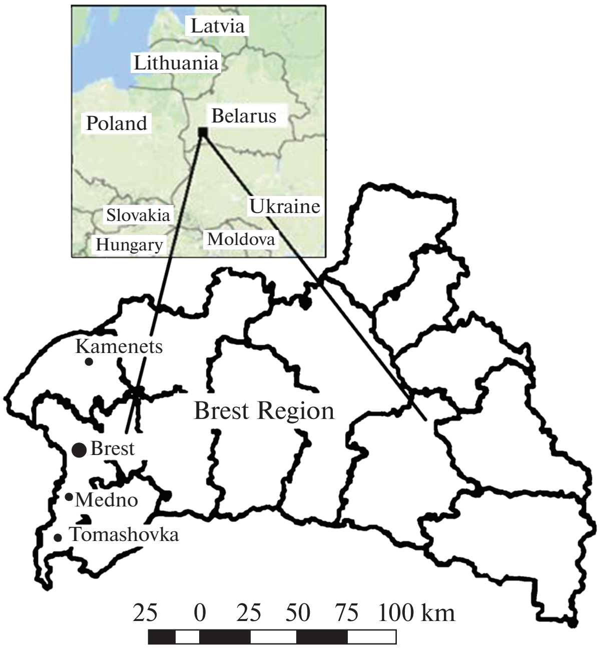

Fig. 1. The scheme of research sites.

of Brest Region of the Republic of Belarus, and to determine the carcinogenic and non-carcinogenic risk for the health of the population as a result of consuming contaminated mushroom products.

The investigation objectives are: 1) to determine the level of contamination of the topsoil, forest conditionally with heavy metals within the boundaries of natural forest ecosystems of contaminated and background territories; 2) to assess the degree of contamination and to find out the peculiarities of heavy metals accumulation in fruit bodies of certain edible mushroom species with either tubular or lamellar hymenophore; 3) to determine the degree of carcinogenic and non-carcinogenic risk for the health of the population from chemical substances which get into the bodies as a result of consuming contaminated macromycetes.

MATERIALS AND METHODS

The studied area and the object of research. The investigations were conducted in 2014–2016 in Brest and Kamenets districts of Brest Region of the Republic of Belarus. The research sites included forests adjoining Brest-Moscow M-1/E-30 highway exposed to the intensive anthropogenic impact (Brest region), as well as the forests within the boundaries of Belovezhskaya Pushcha National Park (Kamenets district), which belong to non-contaminated conditionally background territories (Figure 1).

The forests of the studied area is represented by pine, birch and oak plantations. The sod-podzolics (without subdivision) soils are predominant in the soil mantle of the contaminated and background territories (Umbric Retisols, WRB, 2014) (WRB, 2014).

The objects of the research were edible mushrooms of six species: *Leccinum aurantiacum* (Bull.) Gray, *L. scabrum* (Bull.) Gray, *Suillus granulatus* (L.) Roussel, *S. variegatus* (Sw.) Richon, *Cantharellus cibarius* Fr., *Russula xerampelina* (Schaeff.) Fr.

Sampling and analysis of soil and forest duff. The soil samples were taken from July to September in accordance with the requirements of ISO 10381-4:2003, IDT. They were selected from the sites of 50 × 200 m. One combined soil sample consisted of 8 to 10 point samples. Two types of soil samples were selected (taken): the upper organic-mineral horizon (forest duff) 0–3 см and mixed sample of the upper soil horizons 0–15 см. The acid soluble (1N HCl extractant) and exchange (extractant – buffered ammonium and acetate extract from pH 4.8) forms of Zn, Cu, Mn, Pb, Cd, Ni and Co in soil samples and forest conditionally were determined by the method of atomic and absorption spectrometry on SOLAAR MkII-M6 Double Beam AAS device.

Selection and analysis of mushrooms. Both the soil and forest conditionally samples and the mycological produce were collected from the same sites from July to September. Every mixed sample contained at least 3 fruit bodies. In total, 63 mixed samples of mushrooms were selected and analysed, including carpophore, hymenophore and stipes. All collected fruit bodies of macromycetes were mechanically cleansed from dirt with a plastic knife, rinsed with distilled water, cut into pieces and dried in the oven at the temperature of 65°C reaching air-dry state. Then the samples were reduced to fragments in the laboratory mill and sifted through a sieve with 1 mm diameter mashes. The residues on the sieve were ground in the agate mortar, then added to the sieved part and thoroughly mixed. The mineralization of mushroom samples was done through dry ashing in accordance with the standard 26929-94. Acidic extraction of heavy metals from the ash was carried out with a dilute 1 : 1 HNO_3. The concentration of heavy metals was determined by the atomic absorption spectrometer with a flame atomizer SOLAAR MkII-M6 Double Beam AAS in accordance with the standard 30178-96.

Features of accumulation of heavy metals. The biological absorption coefficient (BAC) was used to analyse the ability of the species of macromycetes to accumulate chemical elements. It was calculated as the ratio of the content of the element in the fruit bodies of mushrooms to its background content in the soils of Brest region (Matveev, Bordon, 2013).

The indicator of element biotics (IEB) (Glazovskiy, 2006) was used to estimate the biogeochemical connection between the chemical composition of a living organism and the biosphere. It was calculated as the ratio of the content of the element in fruit bodies of mushrooms to its clark in the lithosphere (for Belarus) (Matveev, Bordon, 2013).

The accumulation coefficient (AC) was used to characterize the peculiarities of the transition of chemical elements from substrate to fungi. It was calculated as the ratio of the content of the element in fruit bodies of mushrooms to the concentration of exchange and acid-soluble forms of chemical elements in the forest conditionally (Ivanov et al., 2008).

The hazard ratio of the heavy metals content (C_{haz}) was used to assess the level of contamination of macromycetes. It was calculated as the ratio of the average content of the element in the fruit body to the value of its maximum allowed concentration.

Assessment of non-carcinogenic and carcinogenic risk. The assessment of the degree of non-carcinogenic and carcinogenic risk was carried out according to conventional techniques (Onishchenko et al., 2002; R 2.1.10.1920-04, 2004). The value of the hazard quotient of the heavy metals content (HQ) in fruit bodies of separate species of macromycetes was determined for each element investigated as a quotient from dividing the daily intake of the element into its reference dose. The total hazard index (HI) was determined as the sum of hazard indices of each separate heavy metal. The assessment of the carcinogenic risk was made by calculating the individual and total risk. The individual carcinogenic risk was determined as the multiplication of the daily intake of the element in the body to the factor of incidence of carcinogen, whereas the total risk was determined as the sum of individual risks identified for each carcinogens. When determining the risk, we considered the fact that the season of wild growing mushrooms consumption lasts 6 months (from May to October). The daily consumption of mushrooms amounts to 0.143 kg of fresh weight for adults and adolescents over 14 years old and 0.071 kg – for children and adolescents under 14 years old (EFSA Journal, 2012). The average body weight of the representatives of various age groups – children at the age of 10–14 years, adolescents of 14–18 and adults – amounted to 43 kg, 61 kg and 70 kg respectively (Zsigmond et al., 2015).

Microsoft Excel 2013 and Statistica 12.0 applied program package were used to process experimental data.

RESULTS AND DISCUSSION

Heavy metals in soil and forest conditionally. The general specific feature of the investigated territory is that the content of both exchange and acid soluble forms of Zn, Cu, Mn, Ni and Co is higher in the forest duff than in 0–15 cm soil layer, which is a common feature of all soils in the temperate zone forests (Table 1).

It is necessary to note, that the main sources of soil pollution with heavy metals in Brest region are emissions from vehicles, energy facilities and industrial enterprises (Kakareka, Kuharchik, 2012). Besides, special geographical location of Belarus introduced a transboundary component in atmospheric deposition on its territory (Mikhalchuk, 2017). In particular, the amount of lead participating in a transboundary transfer and falling into the territory of Belarus, only from the closest developed western countries (Poland, Germany) and Russia can be at least 355 tons per year, cadmium – more than 15 tons per year (Tonchevski, 2015).

The forest duff within the boundaries of the conditionally background territory proved to be more contaminated with such elements as Zn (3.18 MPC) and Cu (3.08 MPC), whereas the content of Mn exceeded the maximum allowed concentration both in the soil and in the forest duff by 1.8 and 2.0 times respectively. The forest duff within the boundaries of the technogenous-contaminated areas also contained manganese in quantities that on average exceeded the threshold concentration by 1.5 times. The above results are connected not only with the effects of the technogenic factors, but mainly with the biophilic factors, they are active in the biological cycle and accumulate quite intensively in the forest topsoil. A comparatively high content of manganese is the result of its biological accumulation in conditions of a very long interaction of forest vegetation with the soil, which is very typical of the indigenous forests of Belovezhskaya Pushcha (Mikhalchuk et al., 2016).

Besides, it is common knowledge that woody vegetation participates actively in the podzol paedogenesis formation and in the redistribution of nutrients and microelements along the soil profile. In particular, oak leaves have considerable concentrations of manganese (BAC = 4.71), fresh forest duff and young needle litter accumulate significant amount of copper (BAC = = 3.35–5.97), *Betula pendula* (L.) Roth has considerable concentrations of zinc, its accumulation coefficient in leaves and young branches amounts to 5–7 units. It is this fact that explains why the content of Zn, Cu and Mn in the forest conditionally within the conditionally background territory, which is presented by pine, oak and birch stand exceeds the content of these elements in the forest conditionally on technologically contaminated territory (Myslyva, Bilyavskiy, 2016).

The content of exchange and acid soluble forms of lead in the soil cover of the contaminated area is higher than in the forest duff. A high content of lead in the soil in the investigated area proved to be of the technogenic nature, and it is predetermined, in the first place, by the lateral intake of contaminated aerial masses by forest ecosystems and flood runoff from motor transport landscapes bordering on these ecosystems or just crossing them. Similar regularities were also found in the investigations carried out by Ukrainian scientists (Myslyva, 2013). The source of cadmium emission in the soil within the polluted and conditionally background areas exceeds maximum allowable concentration on average by 1.2 times and the index of background content for Brest region by 5–6 times. It is the leaf and needle litters, which absorb this pollutant from the contami-

Table 1. The heavy metals content in forest duff (depth 0–3 cm) and mixed sample of the upper soil horizons (depth 0–15 cm) in forest ecosystems on the territory of Brest Region of the Republic of Belarus

Sampling depth and number of samples	Content of element, мg/kg air-dry mass						
	Zn	Cu	Mn	Pb	Cd	Ni	Co
Technogenous polluted territory							
Forest duff, 0–3 cm, $n = 12$ pH = 3.6–3.8	12 ± 0.6 / 17 ± 0.9	1.3 ± 0.1 / 1.5 ± 0.1	89 ± 5 / 203 ± 11	1.3 ± 0.1 / 3.7 ± 0.2	0.11 ± 0.01 / 0.14 ± 0.01	0.10 ± 0.01 / 0.35 ± 0.2	0.11 ± 0.05 / 0.48 ± 0.3
Umbric Retisols, 0–15 cm, $n = 12$ pH = 4.9–5.1	0.42 ± 0.1 / 0.89 ± 0.04	0.63 ± 0.05 / 1.12 ± 0.1	15 ± 0.8 / 35 ± 2	3.17 ± 0.2 / 6.33 ± 0.3	0.5 ± 0.03 / 0.6 ± 0.03	0.05 ± 0.03 / 0.14 ± 0.08	0.05 ± 0.03 / 0.32 ± 0.2
Conditionally background territory							
Forest duff, 0–3 cm, $n = 9$ pH = 3.8–4.0	73 ± 4 / 104 ± 5	9.5 ± 0.5 / 17 ± 0.9	120 ± 7 / 273 ± 15	3.1 ± 0.2 / 8.9 ± 0.5	0.5 ± 0.03 / 0.7 ± 0.04	1.1 ± 0.05 / 3.8 ± 0.2	0.53 ± 0.03 / 1.5 ± 0.08
Umbric Retisols, 0–15 cm, $n = 9$ pH = 5.1–5.3	5.7 ± 0.3 / 8.0 ± 0.434	0.14 ± 0.01 / 1.3 ± 0.1	109 ± 6 / 249 ± 13	1.5 ± 0.1 / 4.2 ± 0.3	0.6 ± 0.03 / 0.8 ± 0.04	0.10 ± 0.01 / 0.34 ± 0.02	0.15 ± 0.01 / 0.71 ± 0.04
MAC (GN 2.1.7.12-1-2004; GN 1.7.2041-06)	23.0	3.0	80.0 (acid soluble forms) 400.0 (exchange forms)	6.0	0.5	4.0	5.0
Clark (Matveev, Bordon, 2013)	36.6	16.2	369.3	14.9	3.6	11.9	7.8

Note. Numerator – exchange form, denominator – acid soluble form; MAC – maximum allowable concentration.

nated air masses coming from nearby transport landscapes.

Features of the accumulation of heavy metals by macromycetes. The concentration of chemical substances in the fruit bodies of macromycetes is species and environment-specific (Sazanova et al., 2017), and also depends on the content of chemical elements in parent species, their mineral composition, soil type, its agrochemical, physical and chemical properties as well as on the chemical composition of forest conditionally (Myslyva, Bilyavskiy, 2016). The average content of chemical elements in the studied species of mushrooms was correlated with the data obtained by other researchers, in particular, carried out in similar soil and climatic conditions of Poland. For the contaminated areas, the average zinc content in fruit bodies of macromycetes was 172–270 mg/kg; copper – 26–95; manganese – 5–85; lead – 0.5–2.0; cadmium – 0.2–2.0; nickel – 0.2–1.6; cobalt – 0.4–1.5 mg/kg of dry weight. For macromycetes growing within the conditionally background territories, the average zinc content in the fruit bodies was 75–224 mg/kg; copper – 7–44; manganese – 4–31; lead – 0.1–1.1; cadmium – 0.5–3.0; nickel – 0.3–1.0; cobalt – 0.3–0.5 mg/kg of dry weight (Table 2).

Mushrooms can accumulate heavy metals even when their concentration in the soil is relatively low (Falandysz et al., 2003; Kula, 2011; Sazanova et al., 2017). Under the conditions of forest ecosystems subject to the intensive technogenic effects (highwayside forest stock) and irrespective of macromycetes species their fruit bodies accumulate copper (AC_{BAA} = 20.73–76.37; AC_A = 16.91–62.3) and zinc (AC_{BAA} = 16.04–23.02; AC_A = 11.29–16.20) most intensively (Table 3). The maximum zinc accumulating capacity has shown *Leccinum aurantiacum*. High capacity of this species to accumulate high concentrations of Zn and Cu has also been revealed in investigations carried out in conditions of Ukrainian Polesie (Bilyavskiy et al., 2016), within the boundaries of Yaroslavl Region (Pleshcheevo Lake National Park) (Pelgunov, Pelgunova, 2015) and within the boundaries of Poland (Mleczek et al., 2013b). The highest ability to concentrate both exchange and acid-soluble forms of copper was demonstrated by *Cantharellus cibarius*. Other researchers (Solomko et al., 1986; Alonso et al., 2003) also registered the ability of this species to accumulate Cu.

Copper (AC_{BAA} = 2.65–4.80; AC_A = 1.43–2.60) and zinc (AC_{BAA} = 1.02–3.07; AC_A = 0.72–2.16) accumulated intensively in the fruit bodies of macromycetes growing within the limits of the conditionally background territory. A characteristic feature of zinc is its ability to bind with humic acids with conversion into less mobile compounds – zinc humates. Thus, the

Table 2. The content of heavy metals in the fruit bodies of macromycetes, growing within the technogenous polluted territory (PT) and conditionally background territory (BT) of Brest Region of the Republic of Belarus and neighboring countries

Mushroom species	Content of elements, mg/kg air-dry mass							Source
	Zn	Cu	Mn	Pb	Cd	Ni	Co	
Cantharellus cibarius (PT)	263 ± 47	95 ± 19	85 ± 19	0.81 ± 0.28	0.46 ± 0.12	1.60 ± 0.24	1.52 ± 0.29	Authors' data
C. cibarius	13.2–20.4	13.2–15.3	–	0.04–13.2	0.036–0.19	0.1–2.64	13.2–20.4	Sazanova et al. (2017): Finland, Poland
	95.2–139	13.0–28.4	–	43.7–49.8	2.24–3.37	–	–	Záhorcová et al. (2016): Slovakia
	69–100	33–77	20–63	–	0.49–0.67	–	–	Brzezicha-Cirocka et al. (2016): Eastern Poland
	34.05–69.15	20.69–45.92	16.1–19.5	–	0.17–0.5	0.33–0.79	0.12	Zsigmond et al. (2015): Romania, urban region
	–	34–133	4.3–55.0	0.16–1.0	0.038–0.11	1.5–3.4	0.004–0.26	Koroleva, Ohrimenko (2015): Kaliningrad Region, Russia
	–	–	–	1.71	0.44	2.08	–	Pelkonen et al. (2006): Finland
	140.6	–	–	–	–	–	–	Georgescu, Busuioc (2011): Dambovita county, Romania
	57.5	32.7	–	1.63	1.0	0.83	–	Stankeviciene D (2004): Lithuania
Suillus granulatus (PT)	189 ± 33	30 ± 5	7 ± 1.5	0.57 ± 0.14	0.24 ± 0.06	0.19 ± 0.03	0.51 ± 0.08	Authors' data
	–	37.4–60.6	–	–	–	–	0.06–1.02	Svoboda, Chrastny (2007): rural area, Czech Republic
Suillus species	32.8–128.8	0.06–28.2	27.6–71.7	0.03–27.7	0.15–8.95	0.94–18.3	–	Sazanova et al. (2017): Eastern Poland
S. luteus	78–180	11–33	24–62	–	0.19–0.80	–	–	Brzezicha-Cirocka et al. (2016): Eastern Poland
	44.55	26.33	–	0.78	2.98	1.04	0.28	Mleczek et al. (2013): Poland
S. variegatus (BT)	75 ± 16	7 ± 1.5	4 ± 0.8	2.13 ± 0.48	1.16 ± 0.24	0.26 ± 0.07	0.45 ± 0.07	Authors' data
S. variegatus	90	19	13	0.2	1.0	–	0.07	Szubstarska et al. (2012): Northern Poland
	–	–	–	1.11	1.46	1.17	–	Pelkonen et al. (2006): Finland
	70.5–73.3	–	–	–	–	–	0.15–0.22	Borovička, Řanda (2007): Czech Republic

Table 2. (Contd.)

Mushroom species	Content of elements, mg/kg air-dry mass							Source
	Zn	Cu	Mn	Pb	Cd	Ni	Co	
Leccinum scabrum (PT)	173 ± 35	26 ± 5	11 ± 2	0.53 ± 0.12	1.99 ± 0.45	0.62 ± 0.16	0.42 ± 0.07	Authors' data
L. scabrum (BT)	224 ± 63	25 ± 4	16 ± 3	0.11 ± 0.02	3.24 ± 0.68	0.95 ± 0.17	0.31 ± 0.05	
L. scabrum	–	–	–	0.4	0.8	–	–	Sazanova et al. (2017): Eastern Poland
	140–260	14–39	6.4–43.0	0.78–7.5	1.2–8.8	0.76–0.91	0.011–0.48	Zhang et al. (2013): Sudety Mountains, Poland
	–	22.0–29.6	–	–	–	–	0.09–0.15	Svoboda, Chrastny (2007): rural region, Czech Republic
	15.83–68.33	7.1–15.77	–	0.1–15.4	0.33–2.60		–	Stankeviciene (2004): Lithuania
L. aurantiacum (PT)	271 ± 52	60 ± 14	5 ± 0.8	1.92 ± 0.28	0.68 ± 0.15	0.51 ± 0.07	0.54 ± 0.08	Authors' data
L. aurantiacum	110.37	–	–	0.38–33.8	0.35–2.36	8.51	–	Sazanova et al. (2017): Eastern Poland
	20–320	11–150	5.4–73.0	–	–	–	–	Brzezicha-Cirocka et al. (2016): Eastern Poland
	72.04–162.42	12.65–22.89	–	0.82–1.42	1.96–3.94	1.92–2.99	0.64–1.48	Mleczek et al. (2013b): Poland
Russula xerampelina (BT)	95 ± 15	44 ± 11	31 ± 9	1.14 ± 0.27	0.47 ± 0.09	0.32 ± 0.04	0.37 ± 0.06	Authors' data
R. xerampelina	–	43–79	14–15	0.07–1.3	1.2–4.7	4.2–5.0	0.009–0.03	Koroleva, Ohrimenko (2015): Kaliningrad Region, Russia
	19.7–99.6	–	–	–	–	–	–	Georgescu, Busuioc (2011): Dambovita county, Romania
Russula species	19.3–58.2	10.8–73.0	24.2–26.4	0.77–2.6	0.06–2.0	3.2	–	Sazanova et al. (2017): Eastern Poland
	30.4–146.7	6.6–34.1	11.1–219.2	–	–	0.3–2.1	0.009	Elekes, Busuioc (2013): Targoviste, Romania
	–	25.2–64.0	–	–	–	–	0.29–1.81	Svoboda, Chrastny (2007): rural area, Czech Republic

consolidation of Zn with sandy-loamy soils on the conditioned-background territory occurs, and a less intensive accumulation of it with the fruit bodies of macromycetes takes place. Copper is more mobile in the upper genetic horizons of the soil than zinc, and its maximum mobility occurs in forest conditionally with pH in the range of 3.6–3.8 pH units. For this reason, copper accumulates most intensively in mushrooms that grow within the National Park "Belovezhskaya Pushcha". *Leccinum scabrum* is the absorber of zinc, and *Russula xerampelina* is the absorber of copper. A high ability to accumulate zinc was established for species of genus *Leccinum* in the studies by Mleczek et al. (2013b). The intensive accumulation of copper by *R. xerampelina* is obviously connected with the place where it grows, and with the ability of the given species to accumulate copper, which was highlighted by some scientists (Elekes, Busuioc, 2013; Bilyavskiy et al., 2016).

Every species of macromycetes investigated is characterized by low or extremely low ability to accumulate lead (AC_{BAA} = 0.03–1.51; AC_A = 0.01–0.52). These results also correlate with the results obtained in Finland, which, in particular, revealed a low capacity for lead accumulation by *Leccinum* species (Pelkonen et. al., 2006). A low capacity for lead concentration for *Leccinum aurantiacum* and *Cantharellus cibarius* was demonstrated in the studies carried out on the territory of the Republic of Poland (Mleczek et al., 2013b). The reason for a rather low intensity of lead accumulation by symbiotrophic macromycetes is that this element accumulates in the upper genetic horizons of the soil profile. These horizons are rich in organic matter that binds this element. Lead can be a part of the Fe and Mn oxides, which are abundant in the soil of the studied area, thus becoming inaccessible.

Manganese is the least accumulated element by mushrooms growing on both contaminated and conditionally background territories. Depending on the location and macromycetes species the coefficient of its biological absorption varies within the limits of 0.02 to 0.42 for acid soluble and from 0.04 to 0.95 – for exchange forms. Manganese is the element of weak accumulate and intake for 30% of the species investigated. The above findings correlate with the data obtained under the investigation into the peculiarities of manganese accumulation by macromycetes on Irtysh land (Sibirkina, 2009). *C. cibarius* accumulated manganese most intensively. The manganese accumulation coefficient for this species reached 0.95 for exchange and 0.42 for acid-soluble forms. The ability of this species to accumulate manganese is highlighted in other investigations (Sesli et al., 2008). Since the values of the manganese biological accumulation coefficients do not exceed meaning one, it can be assumed that it enters into the fruit bodies of macromycetes exclusively from the substrate. This statement is also true for the accumulation of lead.

Table 3. Coefficients of accumulation of heavy metals in the fruit bodies of macromycetes

Species	Element	AC_{BAA}	AC_A	BAC	IEB
Technogenous polluted territory					
Leccinum scabrum	Zn	14.66	10.31	12.97	4.71
	Cu	20.73	16.91	11.18	1.59
	Mn	0.12	0.05	0.18	0.03
	Pb	0.42	0.14	0.13	0.04
	Cd	18.09	14.21	19.90	0.55
	Ni	6.20	1.77	0.26	0.05
	Co	3.82	0.88	0.20	0.05
L. aurantiacum	Zn	23.02	16.20	20.38	7.40
	Cu	48.39	39.47	26.09	3.70
	Mn	0.05	0.02	0.08	0.01
	Pb	1.51	0.52	0.47	0.13
	Cd	6.18	4.86	6.80	0.19
	Ni	5.00	1.43	0.21	0.04
	Co	4.91	1.13	0.26	0.07
Cantharellus cibarius	Zn	22.38	15.74	19.80	7.20
	Cu	76.37	62.30	41.17	5.85
	Mn	0.95	0.42	1.42	0.23
	Pb	0.64	0.22	0.20	0.05
	Cd	4.18	3.29	4.60	0.13
	Ni	16.00	4.57	0.67	0.13
	Co	13.64	3.13	0.71	0.19
Suillus granulatus	Zn	16.04	11.29	14.20	5.16
	Cu	23.81	19.42	12.83	1.82
	Mn	0.08	0.03	0.11	0.02
	Pb	0.45	0.15	0.14	0.04
	Cd	2.18	1.71	2.40	0.07
	Ni	1.90	1.36	0.08	0.02
	Co	4.55	1.56	0.24	0.06
Conditionally background territory					
Leccinum scabrum	Zn	3.07	2.16	16.85	6.12
	Cu	2.65	1.43	10.65	1.51
	Mn	0.14	0.06	0.27	0.04
	Pb	0.03	0.01	0.02	0.01
	Cd	6.48	4.63	32.40	0.90
	Ni	0.87	0.25	0.40	0.08
	Co	0.58	0.21	0.15	0.04
Suillus variegatus	Zn	1.02	0.72	5.62	2.04
	Cu	0.76	0.41	3.04	0.43
	Mn	0.04	0.02	0.07	0.01
	Pb	0.65	0.22	0.49	0.13
	Cd	2.32	1.66	11.60	0.32
	Ni	0.24	0.07	0.11	0.02
	Co	0.85	0.30	0.21	0.06
Russula xerampelina	Zn	1.30	0.92	7.16	2.60
	Cu	4.80	2.60	19.30	2.74
	Mn	0.26	0.11	0.52	0.08
	Pb	0.37	0.13	0.28	0.08
	Cd	0.94	0.67	4.70	0.13
	Ni	0.29	0.08	0.13	0.03
	Co	0.70	0.25	0.18	0.05

Note: AC_{BAA} – accumulation coefficient of exchange forms; AC_A – accumulation coefficient of acid soluble forms; BAC – biological absorption coefficient; IEB – indicator of element biotics.

Table 4. Hazard coefficient of heavy metals (C_{haz}) in the fruit bodies of macromycetes

Species	Hazard coefficient of (C_{haz})					
	Zn	Cu	Pb	Cd	Ni	Co
Technogenous polluted territory						
Cantharellus cibarius	13.17	9.47	1.62	4.60	3.20	3.00
Suillus granulatus	9.44	2.95	1.14	2.40	0.38	1.00
Leccinum scabrum	8.63	2.57	1.06	19.90	1.24	0.84
L. aurantiacum	13.55	6.00	3.84	6.80	1.00	1.08
Average meaning	11.20	5.25	1.92	8.43	1.46	1.48
Conditionally background territory						
Russula xerampelina	4.76	4.44	2.28	4.70	0.64	0.74
Suillus variegatus	3.74	0.70	4.00	11.60	0.52	0.90
Leccinum scabrum	11.20	2.45	0.20	32.40	1.90	0.62
Average meaning	6.57	2.53	2.16	16.23	1.02	0.75
MAC, mg/kg	20	10	0.5	0.1	0.5	0.5

Cadmium is most intensively accumulated by *Leccinum scabrum* (AC_{BAA} = 18.09; AC_A = 14.21) and *L. aurantiacum* (AC_{BAA} = 6.18; AC_A = 4.86), growing within the technogenous polluted territory. *L. scabrum* (AC_{BAA} = 6.48; AC_A = 4.63) and *Suillus variegatus* (AC_{BAA} = 2.32; AC_A = 1.66) also differed in their ability to accumulate this pollutant; growing within a conditionally background territory. The investigations of Ivanov et al. (2017) point out that the content of cadmium in fruit bodies of mushrooms increases 4–5 times even with insignificant soil contamination, which is due to high mobility of the given element, and our findings confirm it. The ability of *Leccinum scabrum* to accumulate increased amounts of cadmium is also underlined by other investigations which were carried out in conditions of Ukrainian Polesie (Bilyavskiy et al., 2016), Romania (Zsigmond et al., 2015), and Poland (Mleczek et al., 2013b). It is necessary to note that *L. scabrum* belongs to the *Boletaceae* family which representatives are predisposed to cadmium bioaccumulation. The ability of *Boletus edulis*, *B. badius* and *B. chrysenteron* species to accumulate this pollutant was registered in some investigations (Pelkonen et al., 2006; Mleczek et al., 2013a, 2013b; Zsigmond et al., 2015; Koroleva, Ohrimenko, 2015; Bilyavskiy et al., 2016). The reason of active cadmium accumulation by macromycetes within the limits of conditionally background territory, in our opinion, is its increased mobility because of high values of the soil substrate pH, which amount to 3.8–4.0 pH units. A considerable geochemical affinity between Zn and Cd predetermines similar transfer of these metals to the plants and fruit bodies of macromycetes, which obviously absorb more mobile cadmium instead of zinc. Besides, in many biochemical processes cadmium can replace zinc, since their chemical properties are almost similar.

The content of nickel and cobalt in the substrate did not exceed the critical concentration, though these elements were accumulated in surplus quantities mainly in the fruit bodies of macromycetes growing within highwayside forests (AC_{BAA} = 1.9–16.0; AC_A = 1.36–4.57 for Ni and AC_{BAA} = 3.82–13.64; AC_A = 0.88–3.13 for Co). The nickel and cobalt accumulator was *Cantharellus cibarius*. The ability of species belonging to symbiotrophs, in particular, species of the genus *Suillus* to accumulate cobalt is also indicated by Zsigmond et al. (2015). In general, a higher concentration of Zn, Cu, Cd, Ni and Co in the fruit bodies of macromycetes was observed in comparison with the substrate on which they grow, whereas the content of lead and manganese was higher in the substrate.

Contamination of mushrooms with heavy metals. Despite the fact that soil, and forest floor within the limits of Belovezhskaya Pushcha National Park contain larger quantities of zinc and copper, than soil and forest bedding in the limits of the technogenous contaminated territory, the hazard coefficients of their content in macromycetes, which grow in the limits of conditionally background territory, have the lowest values as compared with such for the technogenous contaminated territory (Table. 4).

Although the soil in the environments of highway contained the quantities of zinc and copper equivalent to only 0.02 and 0.21 MAC respectively, the hazard coefficient of content of the above elements in the fruit bodies of macromycetes reached the values of 8.6–13.6 for zinc and 2.6–9.5 for copper. Within the limits of natural forest ecosystems, they were 3.7–11.2 for zinc and 0.7–4.4 for copper. This fact confirms that a species and ecological environment are the main factors, which influence the accumulation of heavy metals by macromycetes which was also noted by a number of researchers (Sibirkina, 2009; Koroleva, Ohrimenko, 2015; Sazanova et al., 2017; Ivanov et al., 2017).

There is one more fact to consider. The contamination of the macromycetes fruit bodies growing within both contaminated and conditionally background territories with lead is relatively similar. The coefficient of hazard (C_{haz}) of lead content in the fruit bodies of macromycetes growing on the technogenous-disturbed territory varied from 1.1 to 3.8, whereas its value on the territory of natural forest ecosystems amounted to 0.2–4.0 on the average depending on the variety of macromycetes (Table 4).

Table 5. The total hazard index (HI) of the heavy metals content in the fruit bodies of separate species of macromycetes

Species	Age-groups of the population living in the study region		
	10–14 years	14–18 years	adults
Technogenous polluted territory			
Cantharellus cibarius	1.28	1.82	1.59
Suillus granulatus	0.48	0.68	0.59
Leccinum scabrum	1.02	1.45	1.26
L. aurantiacum	1.00	1.42	1.24
Conditionally background territory			
Russula xerampelina	0.69	0.98	0.85
Suillus variegatus	0.59	0.84	0.73
Leccinum scabrum	1.44	2.05	1.78

The similar tendency was observed with cadmium. Besides, it has been revealed that the fruit bodies of *Leccinum scabrum* growing on both contaminated and conditionally background territory had the maximum cadmium contamination equivalent – 19.9–32.4 of critical concentration, whereas the average content of the given pollutants in the soil did not exceed 1.0–1.2 of critical concentration. The maximum contamination with both nickel and cobalt was observed in the fruit bodies of *Cantharellus cibarius*, which shows the ability to accumulate the given pollutants, as it is highlighted in the research of Murati et al. (2015).

Carcinogenic and non-carcinogenic risk. The research has provided the opportunity to determine the risk of the human's intake, including carcinogenic one, because of consuming such species of macromycetes contaminated with heavy metals, which were calculated for various age groups. The risks were assessed according to the value of hazard quotient (HQ) for every metal and the value of the total hazard index (HI) which is determined as a sum of all hazard quotients. If the total hazard quotient does not exceed meaning one (1), the probability of developing hazardous effects in humans at daily substance intake during the lifetime is inessential, and this effect is characterized as admissible. If the total hazard quotient exceeds meaning one (1), the probability of developing hazardous effects in human's increases in proportion to the increase in HI. In macromycetes growing on both the technogenous contaminated and on the background territory the highest hazard quotient was revealed for cadmium (HQ = 0.10–1.32) and copper (HQ = 0.10–1.02) (Table 5).

In terms of the magnitude of non-carcinogenic risk among all species of macromycetes growing on the technogenous-contaminated territory, only *Suillus granulatus* proved to be the safest when consumed as a food product. This species along with *Russula xerampelina* growing within the background territory proved to be the safest as to the level of their contamination with heavy metals.

When assessing the total risk of damage to certain organs and systems of the human body, it has been established that when eating macromycetes that grow both within the contaminated area and on a conditionally background area, the most likely adverse effects can be for gastrointestinal tract and liver (HQ_{cont} = = 1.84–2.62; HQ_{backgr} = 0.67–0.96), and hematopoietic organs (HQ_{cont} = 1.84–2.62; HQ_{backgr} = 1.91–2.71) (Figs 2, 3). Moreover, the risk of negative effects on the hematopoietic organs when eating macromycetes grown on a conditionally background area is higher than for a contaminated area due to higher levels of macromycetes contamination by cadmium. The maximum contribution to the total risk is made by Cu, Ni and Cd. Regardless of the location of growth and the level of contamination of macromycetes, the maximum non-carcinogenic risk is established for the age group of adolescents aged 14–18.

The assessment of carcinogenic risk, which is understood as the probability of the increase in the incidence of neoplasms in humans due to the peroral effects of chemical carcinogens, was done by calculating the values of individual, total and population risks. Studied. Lead and cadmium as substances with a proven carcinogenic effect, were selected among the chemicals (Table 6).

The total carcinogenic risk due to only two identified carcinogens for man – for the contaminated area is $0.1–2.1 \times 10^{-2}$, and for conditionally background it is $0.3–3.4 \times 10^{-2}$. According to the international crite-

Table 6. The total carcinogenic risk at peroral intake of heavy metals consuming polluted macromycetes

Species	Age-groups of the population living in the studied region	
	10–14 years	14–18 years and adults
Technogenous polluted territory		
Cantharellus cibarius	0.03	0.05
Suillus granulatus	0.01	0.03
Leccinum scabrum	0.10	0.21
L. aurantiacum	0.04	0.08
Conditionally background territory		
Russula xerampelina	0.03	0.06
Suillus variegatus	0.07	0.13
Leccinum scabrum	0.17	0.34

rion scale (Onishchenko et al., 2002) the risk is assessed as high unacceptable for production conditions, and requires taking remedial action or decrease.

Eating mycological products that have the following pollution levels: Cd – 8.4–16.2 MAC; Zn – 6.5–11.0 MPC; Cu – 2.5–5.2 MAC; Pb – 8.4–16.2 MAC; Ni and Co – 1.5 MACs throughout life can lead to the increase in carcinogenic and non-carcinogenic health risks. However, when assessing carcinogenic risk, it should be borne in mind that the stochastic nature of the carcinogenic process, a long latent period, the differences in age sensitivity to the effects of carcinogens do not allow accurately predict the timing of development of malignant neoplasms in the population.

CONCLUSION

Our findings revealed that in modern ecological conditions that have developed within the region under investigation, it is necessary to revise the background areas for monitoring the contamination of forest soils. The intensity of accumulation of macromycetes by pollutants and their ability to concentrate or deconcentrate are equally influenced by both the biological features of a particular species of macromycetes and the geochemical specificity of the biotope. It was found that for fungi, growing in pine-birch with the admixture of oak forest plantations, the contamination of fruit bodies with Zn, Cu and Cd is most likely. Mushroom raw materials harvested within both technologically contaminated and conditionally background ar-

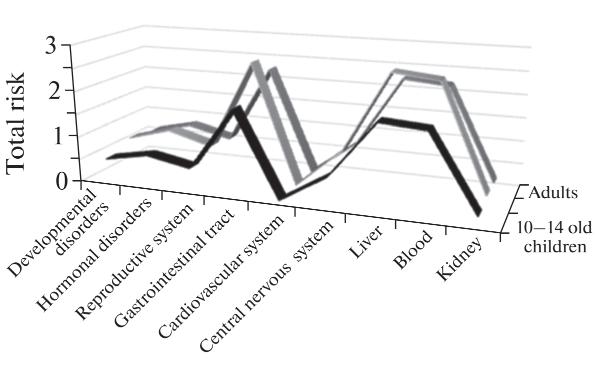

Fig. 2. The total risk of damage to certain organs and systems of the human body when eating macromycetes that grow within a technogenous polluted area, $HQ_{cont.}$

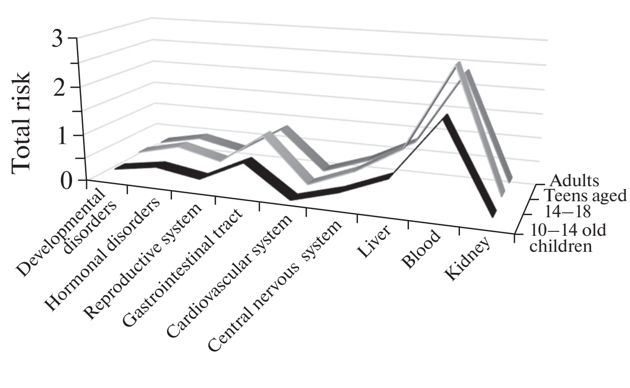

Fig. 3. The total risk of damage to certain organs and systems of the human body when eating macromycetes that grow within a conditionally background territory, $HQ_{backgr.}$

eas are not safe, and its consumption as food can lead to the increase in carcinogenic and non-carcinogenic risks of the incidence of the population.

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АККУМУЛЯЦИЯ ТЯЖЕЛЫХ МЕТАЛЛОВ МАКРОМИЦЕТАМИ В БРЕСТСКОЙ ОБЛАСТИ РЕСПУБЛИКИ БЕЛАРУСЬ

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Исследованы особенности накопления тяжелых металлов различными видами дикорастущих съедобных макромицетов, произрастающими в пределах загрязненных и условно фоновых территорий природных лесных экосистем Брестской обл. Республики Беларусь. Установлено, что в пределах исследованной территории на накопление тяжелых металлов в плодовых телах макромицетов более интенсивно влияют их видовая принадлежность и экологические особенности, нежели характер субстрата и условия места произрастания. Грибное сырье, получаемое в пределах как техногенно-загрязненных, так и условно фоновых территорий Брестской обл., загрязнено тяжелыми металлами, которые по уровню содержания в плодовых телах макромицетов образуют следующий ранжированный ряд: Zn > Cu > > Cd > Pb > Ni > Co. Употребление в пищу грибов, имеющих следующие уровни загрязнения: Cd – 8.4–16.2 ПДК; Zn – 6.5–11.0 ПДК; Cu – 2.5–5.2 ПДК; Pb – 8.4–16.2 ПДК; Ni и Co – 1.5 ПДК на протяжении всей жизни может привести к увеличению канцерогенного и неканцерогенного риска для здоровья. Уровень индивидуального канцерогенного риска от употребления в пищу загрязненных свинцом и кадмием макромицетов составляет $0.1–3.4 \times 10^{-2}$, что по международной критериальной шкале оценивается как высокий, не приемлемый для производственных условий и населения, и требующий принятия мер по устранению либо снижению.

Ключевые слова: загрязнение, канцерогенный и неканцерогенный риск, лесная подстилка, макромицеты, почва, тяжелые металлы