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*HEAVY METALS AND RADIONUCLIDES AS A RISK FACTOR FOR BIOTA IN ENVIRONMENTAL MONITORING OF THE TYUMEN REGION**

SUMMARY. This paper presents the data on the accumulation of heavy metals and radionuclides in the plants (mosses) and animals (voles) in the areas that are intensively subjected to oil extraction, as well as in the background plots under control within the Tyumen Region. The radiation pressure is shown in the industrial regions of West Siberia. It results from the recovery of radionuclides on the surface in these areas and their transfer from the cross-border regions. Heavy-metals pollution and radionuclide contamination as well as radiation pressure pose an additional risk for the development of biological and ecological systems. The indicators of heavy metals and radionuclides accumulation in the organisms of plants and animals are recommended to be used for the Tyumen Region environmental monitoring.

KEY WORDS. Pollutants, heavy metals, radionuclides, ecosystems, monitoring.

Introduction. The contents of heavy metals (V, Ni, Ti, U, Zn, Pb, Cd, Cu, etc.) in the hair, skeleton, etc. [1], [2], [3], [4] et al., taking into account their high physiological significance, is considered one of the indicators in the context of environmental monitoring [5], [6]. In this respect, the special interest is the accumulation of some types of radionuclides in the tissues that can give rise to the local focus of radioactive contamination in the places of synoecium (e.g. cheiropterous animals (*Chiroptera*)) [7] and imply the changes in a number of biochemical processes [8].

In the oil-producing areas of the Tyumen Region the high concentration of a number of elements occurs due to their extraction on the surface with oil or operating activities when producing oil. Definitely, the degree of heavy metal pollution in a deposit area depends on the number and size of spills [9], as well as the oil composition.

Resins and asphaltenes contain the most of oil microelements, including nearly all metals. The total microelement content in oil is hundredths or tenths of a percentage point. Environmentally, micro- and macroelements of oil can be divided into two groups: non-toxic and toxic. The non-toxic and low-toxic elements of oil include Si, Fe, Al, Mn, Ca, Mg, composing much of oil ashes. At the same time, V, Ni, Co, Pl, Cu, U, As, Hg and the other elements, in case of higher concentrations, can have a toxic effect on the biocenosis. In particular, chronic uranium intoxication is characterized by the polytropic effect of uranium on various organs and systems.

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The toxic effects of arsenic and lead are well known. V and Ni (their concentration can reach respectively 0.04 and 0.01% in oil) are the most common among toxic metals. The compounds of these elements in crude oil act as various poisons, suppressing the fermentation activity, affecting the respiratory system, blood circulation, nervous system and skin [10]. The toxic effects of heavy metals are characterized by M.D. Shvaykova [11], and carcinogenicity of some of them is described in the book *Carcinogenic Substances* [12]. Oil usually contains about 98% of hydrocarbons; the rest accrues to the compounds consisting of O, N, S and other microelements [13]. The ratios of these compounds in different types of crude oils can be different; it should be taken into account when conducting monitoring studies in one or another geological and geographical area. For example, aromatic hydrocarbons containing one ring are more common in heavy, tarry, sulfurous oils, that are concentrated in the Central part of the West-Siberian oil and gas area (centered at the city of Surgut), while polycyclic arenes are common in oils with a lower contents of heavy fractions on the area periphery. In terms of naphthenes, the inverse dependence rather takes place [14]. In addition to the effect of oil hydrocarbons on biogeocenoses, it is necessary to take into account the influence on their basic components and microelements that can have significant toxic effects in case of their accumulation in the organism. The contents of many microelements in various oils is known [14], [10], however, their accumulation in the plant and animal organisms in the oil-producing regions is obviously understudied. For example, it is known that vanadium accumulates in the roots, rather than in the aerial parts of the plants growing in the oil-contaminated area [15]. High iron, molybdenum and copper levels are detected in the skeletons and organs of the deer mice, caught in the oil field area [16].

The accumulation of some elements or other in the living organisms depends not only on the biological implication of each element, but is of a species-specific nature, and it depends on the age and sex of small mammals as well [2], [17].

Materials and Techniques. We studied the accumulation of 12 microelements in the aerial part of green mosses and in the skin of northern red-backed voles from the area of the Aganskoe oil field which had been oil-contaminated eight years before (Table 1, 2). The trace analysis was carried out by laser emission microspectrometry using the LMA-10 (CARL ZEISS, JENA) apparatus in the forensic laboratory of the Tyumen Internal Affairs Directorate.

Discussion. The accumulation of Al, Ba, V, Fe, Mn, Ni, Pb, Sr, Ti, Cr in the plants of the oil-contaminated area is shown. The analysis results show that the majority of microelements clearly tends to be accumulated in the aerial parts of green mosses in the contaminated area 2.5-4 times as much as in the control, and for strontium, it is 6 times. These differences are significant for tin, strontium and chromium.

The major part of microelements (Al, Ba, Fe, Mn, Cu, Ni, Sn, Pb, Ti, Cr) tends to be accumulated in the skin of red-backed voles from the oil-contaminated area, and their level of iron, copper, nickel, lead and chromium is even 2.5-4 times as high as in the control. Thus, the animals of the oil spill area significantly differ from the control ones in the content of heavy metals. While the animals of the oil-contaminated area have a lower level of vanadium and strontium (vanadium level in the plants of the control is much lower); there is no acceptable explanation yet [5].

Table 1

Microelement contents in the aerial part of the green mosses in the oil-contaminated area and in the ground control point (ashes, mg/kg)

		Al	Ba	V	Fe	Mn	Cu	Ni	Sn	Pb	Sr	Ti	Cr
Control	X	330	233	0.17	517	367	50	1.7	0.27	5.7	333	76.7	2.7
	m	33	133	0.09	235	66.7	1.2	0.3	0.15	1.3	333	23.3	0.9
Oil	X	730	1667	5.55	1500	467	18.3	6.0	1.17	20	2000	167	9.3
	m	384	726	4.75	287	120	6.0	2.1	0.17	5.8	500	66.7	0.7
Significance of differences	Transcription factor	1.04	1.94	1.12	2.64	0.73	2.17	2.04	4.07	2.42	2.78	1.27	5.97
	P<	—	—	—	—	—	—	—	0.05	—	0.05	—	0.01

Table 2

Microelement contents in the skin of northern red-backed voles in the oil-contaminated area and in the ground control point (ashes, mg/kg)

		Al	Ba	V	Fe	Mn	Cu	Ni	Sn	Pb	Sr	Ti	Cr
Control	X	809	133	11	453	114	7	1	0.3	13	117	104	2
	m	95	84	10	24	93	2	0.3	0.1	2	19	21	1
Oil	X	2000	183	1	1667	300	30	4	0.5	30	50	117	5
	m	500	17	0	167	0	0	0.6	0	0	0	33	0
Significance of differences	Transcription factor	2.34	0.58	1.0	7.20	2.00	11.5	4.47	2.00	8.50	3.53	0.33	3.00
	P<	—	—	—	0.01	—	0.001	0.05	—	0.01	0.05	—	0.05

Thus, there is the coupling accumulation of all the studied elements, except strontium, in the aerial part of the green mosses and in the skin of the northern red-backed voles in the oil-contaminated area; however, the correlation between these indicators is reduced as a result of contamination in comparison with the conditionally background area (Fig. 1).

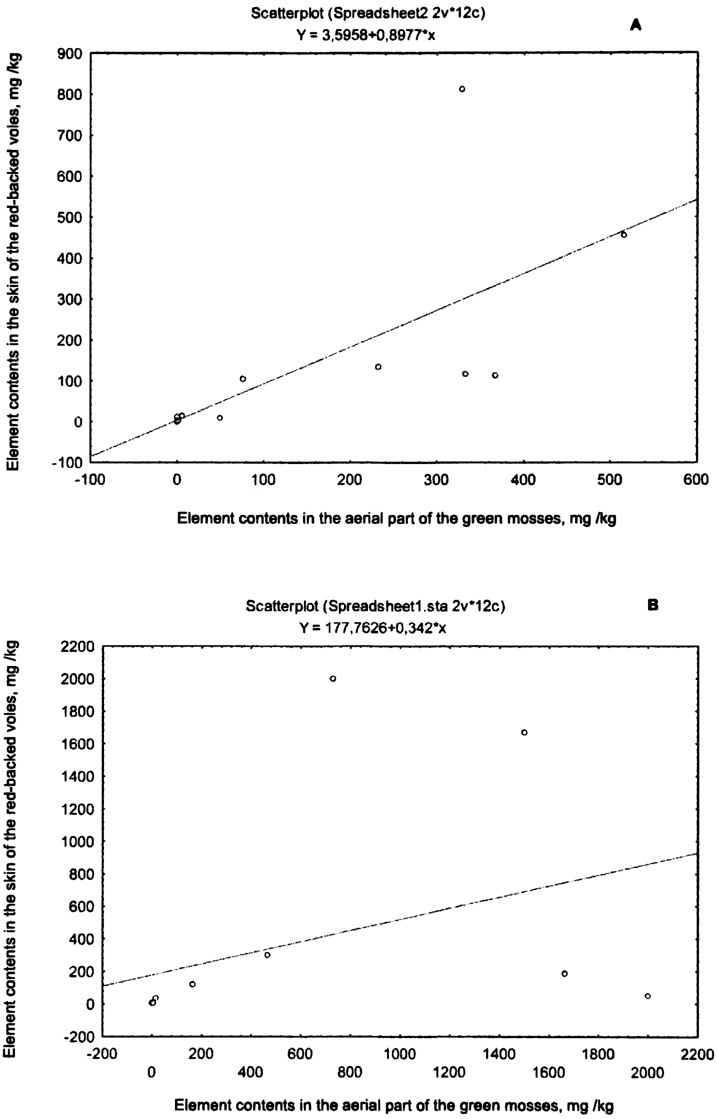

Fig. 1. Dependence of the test element content in the skin of the red-backed voles on their content in the aerial part of the green mosses in the conditionally background area (A) and in the area oil-contaminated eight years before (B)

The presence of uranium as a natural radionuclide in the oil (the content of which in different types of oils varies within two orders: n x 10^{-4} — n x 10^{-2} %, i.e. up to 10-15 mg per 100 g of oil), as well as strontium-90 and caesium-137 as radionuclides, the content of which is mainly caused by human activities (involved in the metabolic processes, they are accumulated in the food chain), correlated with the resinous-asphaltenic fraction [10] or edge waters [18], in some cases it can result in the increase of the total radioactive background in the oil-contaminated area in comparison with the ground control point. Thus, our research shows that the total level of γ and β-radiation in the oil spill areas of the middle Ob is 17.9±0.5 mR/h, which is significantly above the average ($P < 0.001$) within the oil field, where it is 14.5±0.3 mR/h. But the radiation level in the areas of the flares is by 43% higher than the environmental level [19]. Saline and sludge deposits in the well mouths containing naturally-occurring radionuclides, as radium-226, thorium-232 and potassium-40, determine EDR of γ-radiation from 60 up to 5,600 mR/h [20]. The isotopic determination of soils reveals an evident excess of ^{234}U against ^{238}U that is explained by the input of the former by the formation waters. The uranium accumulation in the aerial parts of different species of plants is specified, and the higher concentrations of ^{238}U in comparison with ^{234}U are observed [18]. The family of the former include 19 parent and daughter radionuclides, of which ^{214}Pb and ^{214}Bi account for 98% of the γ-radiation energy in the uranium family. The whole beds of the willow herb (*Epilobium angustifolium*) with absolutely white flowers are registered within the Aganskoye, Vakhskoye and Vatinskoye oil fields; that provides the evidence of the anomalous uranium content in the soils of these areas. The accumulation of radioactive isotopes in vegetation results in their accumulation in the animal bodies, especially in small mammals inhabiting the areas of increased radioactivity. B.V. Testov and A.I. Taskaev [18] point to the considerable U accumulation in the ground and supporting tissues of the root voles. Chronic uranium intoxication is characterized by the polytropic effect of uranium on the various organs and systems. When hardly soluble uranium compounds have been coming into the organism for a long period of time and the biological effect of uranium as an α-radiator is observed, chronic radiation sickness may develop [21]. Along with the major pathology of various organs and systems, renal disorder is predominant in the clinical picture of uranium poisoning. In case of inhalation effects of various uranium compounds (e.g., radium-226 and especially radon-222), which emit the highest dose of radiation [18], frank symptoms of pulmonary pathology are observed. In addition to uranium, a thorium-232 series is of great importance, the γ-radiation intensity of which is distributed among the isotopes of thallium-208 (60%), actinium-228 (25%), lead-212 and bismuth-212 (15%) [22]. Due to carrying up a number of natural and anthropogenic radionuclides (radium-226, thorium-232, potassium-40, cesium-137, strontium-90, etc.) being carried up to the daylight surface near the well mouths and in the places of oil sludge accumulation in the oil field areas, EDR of gamma-radiation reaches 60 up to 5,600 mR/h (with the background of 9-11 mR/h and the permissible dose of 30 mR/h) [23]. Besides, it is reported about the cases of natural increase in the radiation background up to 48-52 mR/h in the geopathogenic zones.

We obtained the data on the vegetation contamination by artificial long-lived radionuclides in different areas of the Tyumen Region. The data were collected in

2001 from 13 observation points combined into six sampling plots that are characterized by the anthropogenic load of various extents (Table 3). The moss-lichen cover, bark and branches of trees were harvested. The chemical testing of the samples for ^{90}Sr and $^{137}C3$ radionuclides was carried out by M.G. Nifontova (Institute of Plants and Animals Ecology, Ural Branch of the Russian Academy of Science, Yekaterinburg). The results of the analyses are shown in Table 3. The sampling plots are given in the table in order of the increase in the anthropogenic load. It occurs due to oil field development for all sampling plots except Tyumen. The relatively poor development of the territory is characteristic of the north-eastern part of the region (Chertovskaya Lake System). The natural complexes of the middle Ob (the city of Nizhnevartovsk), where the region's largest extensive producing oil fields are located, are under the greatest load.

Table 3

Contents of radionuclides in plant samples in different parts of the Tyumen Region (dry mass, Bq/kg)

№	Location	Plant	^{90}Sr	^{137}Cs
1	Chertovy Lakes (Krasnoselkupsky District)	Lichen *(Cladina stellaris, Cladina rangiferina, Cetraria islandica)*	35.0±5.00	80.0±10.00
2	Settlement of Ugut (Surgutsky District)	Lichen *(Cladina stellaris, Cladina rangiferina)*	42.2±4.01	102.2±11.40
3	Muravlenko (Purovsky District)	Lichen *(Cladina stellaris, Cladina arbuscula)*	50.0±10.00	95.0±25.00
4	Settlement of Punga (Sovetsky District)	Lichen *(Cladina rangiferina)*	50.0±10.00	120.0±30.00
4	Settlement of Punga (Sovetsky District)	Sphagnum moss *(Sphagnum sp.)*	55.0±5.00	145.0±25.00
4	Settlement of Punga (Sovetsky District)	Green mosses *(Pleuruzium schreberi, Dicranumpolysetum)*	60.0±20.00	120.0±50.00
4	Settlement of Punga (Sovetsky District)	Polytrichum mosses *(Polytrichum commune, Polytrichum s trie turn)*	105.0±15.00	200.0±50.00
4	Settlement of Punga (Sovetsky District)	Bark and branches of the trees (pine-tree, aspen, birch)	43.3±8.82	93.3±18.56
5	The city of Nizhnevartovsk	Lichen *(Cladina stellaris, Cladina arbuscula, Cladina rangiferina, Peltigera canina)*	62.5±7.50	220.0±56.57
5	The city of Nizhnevartovsk	Bark and branches of the trees (pine-tree, birch)	40.0±10.00	50.0±10.00

6	The city of Tyumen	Lichen *(Cladina stellaris, Cladina arbuscula, Cladina rangiferina)*	40.0±5.77	146.7±31.80
		Green mosses *(Pleurozium schreberi, etc.)*	90.0±30.00	380.0±60.00
		Polytrichum mosses *(Polytrichum sp.)*	70.0±20.00	650.0±90.00
		Bark and branches of the trees (pine-tree, birch)	30.0±10.00	230.0±30.00

The peculiarity of the anthropogenic impact on the biocenosis in the southern area of the region (the city of Tyumen) is caused by the developed agro-industrial complex. Plants, especially the moss-lichen cover, often are indicators of radioactive contamination of the area. The levels of radioactive contamination in the lichens and mosses are mainly determined by intake of the long-lived radionuclides as a part of global fallout in the Ural-Siberian region [24].

However, the data from the table show that the contents of the examined radionuclides in the lichens within the oil-rich areas in the north of the region have a well-defined tendency to increase in proportion to the growth of the anthropogenic load. Thus, the content of ^{137}Cs in the lichen nearby Nizhnevartovsk is 2.8 times and ^{90}Sr is 1.8 times as much as the same indicator in Chertovy Lakes. At the same time, the ratio of $^{137}Cs/^{90}Sr$ radionuclides changes in the plant samples due to the increase of cesium accumulated by the plants. In case this ratio ranges from 1.9 to 2.4 units in the most parts of the investigated areas, near the city of Nizhnevartovsk, it is 3.5 units that is more than 2 times as much as the indicator value in the global atmospheric fallout amounting to 1.6-1.7 units. [25]. The increased values of the lichen cover contamination by artificial radionuclides nearby Nizhnevartovsk can be related to a series of the underground nuclear explosions in the middle Ob area in the early 1980s for dip shooting and enhance oil recovery. When one of the explosions occurred (the Sredne-Balyksky Oil Field), there was a powerful emission of radionuclides to the surface, and the area was contaminated [26]. Nowadays the objects, formed due to the underground explosions, have no owner, and nobody controls the possibility of radioactive gases emission on the surface through the unpressurized boreholes. According to the data from the Nizhnevartovsk Environmental Protection Committee, there is accumulation and concentration of radionuclides in other species of plants and fungi in the area. Thus, the sanitary control of a dried mushroom lot, picked in the area of the Chakhlomey River arm, showed the concentration of radioactive 137 Cs isotopes being 1,542 Bq/kg, that is 2.6 times as much as the permissible level.

When comparing the explored plant samples taken from one geographical point, as the general trend, the most intensive accumulation of radionuclides is registered in the mosses, the least intensive accumulation is in the bark and branches of the trees, the published data confirm this. The mosses of different taxonomic groups also differ internally in the accumulation of radionuclides intensity. The highest rates of ^{90}Sr and ^{137}Cs accumulation are observed in the polytrichum mosses.

The peculiarity of the southern areas of the region is higher accumulation rates of ^{137}Cs in all flora resources in comparison with the northern areas. Thus, the ^{137}Cs

content in the green and polytrichum mosses and bark near the city of Tyumen is higher than the corresponding values in the northern territories (the settlement of Punga) by 3.2, 3.3, 2.5 times, respectively. The ^{137}Cs / ^{90}Sr ratio in the plant samples near Tyumen ranges from 3.7 units in the lichens to 9.2 units in the polytrichum mosses; that indicates the presence of active contamination source in the area in addition to the global atmospheric fallout. Such sources are likely to be the emergencies at the nuclear complexes of the federal subjects nearby the Tyumen Region: Mayak Industrial Association in the Chelyabinsk Region (August, 1993), Beloyarsk Nuclear Power Plant in the Sverdlovsk Region (October, 1993). The backwash of the «eastern» trace of the Chernobyl fallout that had the direction of Chernobyl-Penza-Yekaterinburg-Tyumen is also possible [27].

The increased contents of radionuclides in the oil-producing areas of the Tyumen Region undoubtedly contribute to the increase of the background radiation. Along with endogenous causes of aggravation of the radiation situation, we should consider its exogenous component caused by the technogenic flows (both air and water) from the cross-border regions, which supply the radioactive contamination from the enterprises and polygons: Mayak Industrial Association, Chelyabinsk-65, Siberian Integrated Chemical Plant in Tomsk-7, Yekaterinburg Uzel (Malyshevsk Mine Group), Beloyarsk Nuclear Power Plant, Ural Electrochemical Plant in Yekaterinburg, Plant-44 and Plant-45 in the city of Sverdlovsk), uranium enrichment plants in Novosibirsk, nuclear test polygons in Semipalatinsk-21 and Novaya Zemlya, etc. [28]. Besides, the southern administrative districts of the region are known for the disposal of radioactive wastes, delivered from North Kazakhstan, which are the sources of the secondary radioactive contamination. The construction materials, including natural radionuclides in increased amounts, represent a certain danger to the biota.

All this dictates the urgent necessity to study radiation environment not only in the Tyumen Region, but also in the border areas, and it also makes necessary to carry out special monitoring studies on radionuclides accumulation in the flora and fauna (plants, animals and microorganisms), which substantially contribute to the general radiation background of the region.

In 1988-1998 we carried out the radiation research in the tundra, forest tundra, north, middle, and south tundra zones of West Siberia [29], [30], [31], which included both the background biotopes, and the landscapes that were exposed to anthropogenic influence in a varying degree (construction, oil and gas production, emergency situations in specialized production, etc.). γ and β-radiation levels were detected by the DP-5V radiometer-R-meter, DBGD «Master-1» dosimeter and RM-1203 R-meter. In total, over 1,000 measurements were performed.

The research has shown that the background level of γ-radiation in the investigated natural zones and subzones of the Western Siberian Lowland averages respectively 8.2±0.16, 7.1±0.32, 6.8±0.30, 10.0±0.50, 8.9±0.15 mR/h. According to the published data, the background level of γ-radiation in the sub-taiga and forest-steppe zones of the region is 6-15 mR/h [28], and it is higher in the area adjacent to the Ural mountains: the background EDR is 24 mR/h in the area of Krasny Kamen' railway station [32]. Thus, the relative homogeneity of the background radiation in the region and its conformity to the average level within the country can be certified.

However, it should be noted that various anthropogenic transformation of landscapes in different natural zones of the Western Siberian Lowland results in the significant local increase of the γ-radiation level.

The most significant differences are specified in the «trace» zone of the accident occurred on April 6, 1993, at the Radiochemical Plant of the Siberian Chemical Complex in the city of Tomsk-7 (south taiga). Even three years after the accident, the γ-radiation level of the «trace» remains significantly (at $P<0.001$) 2.5 times (in the soil) and 1.4 times (in the air) as much as the background level. With that, the average radioactive indicators of different kinds of wood-destroying fungi and wood are similar against each other and γ-radiation level in the air. Thus, the version of the radionuclides accumulation in palisada fungi is not confirmed, but it does not apply to pileate fungi, the level of radionuclides accumulation in which is proved (the radiation level is 6 times as much as it is in the soil). However, within the «trace» there is a significantly greater variation (by 1.4 times at $R<0.05$) of the radioactivity of the unfertile formation of *Innonotus obliquus* (cinder conk) in comparison with basidia of another stem parasite — *Phellinus igniarius*. This may indicate the possibility of radionuclides accumulation by unfertile formations of fungi that seem to perform excretory functions. This possibility should be taken into account when preparing chaga as crude drug [33], [34].

The significant increase of γ-radiation level is specified in the oil producing areas in the middle Ob area (middle taiga) where many radionuclides are carried up on the daylight surface together with the oil: if EDR averages 14.5±0.30 mR/h (it is almost 1.5 times as much as the background) in the area of the Samotlor oil field, then in the oil spill areas it is 17.9±0.50 mR/h, that is significant (at $P<0.001$) by 23.4% higher than in the oil field on average. Even more significant increase of the γ-radiation level, as stated before [19], is specified in the zone of the (delete?) oil and gas flares, where it is by 43% higher than the average level within the oil field.

A slight increase of EDR is specified in the cities of the region: e.g., the level of γ-radiation is 8.2±0.23 mR/h in the Central Park, the city of Nadym, and it is only 7.1±0.32 mR/h in the adjacent forest-tundra. The same regularity is observed even in the settlements: EDR is 7.4±0.23 mR/h in the settlement of Yuilsk (north taiga), while in the area of Numto Lake in the same sub-zone it is only 6.8±0.30 mR/h. Harder anomalies are a characteristic of Tyumen and a number of northern towns in the places of paving and in the building spaces, in construction of which the materials containing smelter slag have been used (EDR of γ-radiation is up to 35-55 mR/h [35]).

The results of the radiation research on the Yamal Peninsula can indicate some increase in the γ-radiation level above the background in the areas of gas condensate fields, where EDR is 10-16 mR/h [32]. In the area of the Obskaya-Bovanenkovo railroad construction (southern tundra), a slight (1.1 times) but significant (at $P<0.05$) increase in the level of γ-radiation which averages 8.8±0.19 mR/h. However, the significant increase of EDR and its variability takes place in the construction area in comparison with the background: the variation coefficient amounts to respectively 37.5±2.1% and 34.6±1.9%. All this can certify the increase not only in the level of radiation under the anthropogenic conditions, but also its variability, and this, in return, dictates a need for a larger number of measurements in order to obtain the representative material.

Conclusion. Thus, the components of the biota (lichens, mosses, small mammals, etc.) inhabiting the oil-contaminated areas are subjected to the extra effect of heavy metal accumulation including radionuclides, as well as the increased α, β, γ-radiation, and they should be used in the system of the regional environmental monitoring.

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