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RADIONUCLIDES VARIATION IN MACROLICHENS IN ESTONIA AFTER THE CHERNOBYL ACCIDENT

Abstract. Radioactive pollution from the Chernobyl NPS reactor accident has wide-scale impact through radionuclides fallout over large areas. We used macrolichens belonging to the *Cetraria* and *Cladina* genera for the investigation of ^{137}Cs and ^{90}Sr fallout and migration in the system "plant — soil". Systematic field collections were made in the Rumpo Botanical Sanctuary on Vormsi Island (West Estonian Archipelago Biosphere Reserve) and in Koljaku Reserve (Lahemaa National Park, LNP) during 1986—89, additional data for comparison were collected in the Caucasus, Spitsbergen, Yamal Peninsula, the Urals and Baikal Lake Reserve, and from various regions of the European part of the USSR.

The maximum concentrations of radionuclides of caesium and strontium in macrolichens exceeded those known from literature for the Arctic areas during the period of nuclear testing. In 1986 the highest concentration of ^{137}Cs in Estonia — 6.2 kBq/kg was measured in the *Cetraria islandica* in LNP. In Rumpo Sanctuary the highest concentrations of caesium radionuclide were estimated in July 1986 — 4.5 for *Cl. rangiferina* and 4.4 kBq/kg for *C. cucullata.* Radiocaesium content decreased rapidly in the following years. The highest rate was established for *Cl. rangiferina* — 12.5 in three years. The absolute values of radiostrontium content in the four investigated lichen species before and after the Chernobyl accident do not differ considerably. The decrease of ^{90}Sr concentration is more evident for *Cl. rangiferina* — from 62 in 1986 to 15 Bq/kg in October, 1988. The same trend is obvious for the radionuclides store: in July 1986 the store of ^{137}Cs in the lichen cover was maximum, 1.7 kBq/m^2, and then decreased continuously reaching 0.23 kBq/m^2 in 1989. The highest store of caesium in soil radionuclide was reached in October 1987 — 4.1 kBq/m^3 × 0.02.

Key words: radionuclides, lichens, Estonia.

Introduction

Lichens have long been known as good accumulators of metal cations (Lounamaa, 1956). This is why they are used as natural monitors showing the extent of metal pollution in air and precipitation. There exist a lot of publications which give evidence about this (Мартин, 1985). In radioecology lichens have been known as subjects of research already from the 1960s when nuclear tests were carried out in the atmosphere. In those years considerably high concentrations of radionuclides in the food chain "lichens — reindeer — man" were discovered in the Arctic areas (Borman et al., 1958; Palmer et al., 1963; Miettinen et al., 1963; Watson et al., 1964; Liden, 1961; Liden, Gustafsson, 1966; Hansson, Eberhardt, 1971; Persson, 1971; Martin, Koranda, 1971; Gorman, 1959; Нижников et al., 1969). In the above-mentioned chain lichens were the most important radionuclide depot (Mattson, 1972).

Soon after the accident at the Chernobyl Nuclear Power Station evidence about the moving of air masses containing radioactive elements and their fallout in Northern Europe became known (Persson et al., 1986), which allowed us to assume that the Baltic Sea region would also be under the impact of radionuclides from the air. In addition to investigating the dispersal of radioactive components from Chernobyl in various natural environments (Израэль et al., 1987; Израэль et al., 1988; Higuchi

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et al., 1988; Roy et al., 1988; and others) also the content of radionuclides of caesium and strontium in lichens was studied (Seaward et al., 1988; Papastefancu et al., 1989; Rissanen et al., 1989; Нифонтова, Куликов, 1990). For the territory of Estonia, data on the accumulation of ^{137}Cs in mushrooms (Parmasto, Liiva, 1988) and preliminary data on the dynamics of ^{137}Cs and ^{90}Sr in macrolichens (Martin et al., 1989) exist.

Materials and Methods

Field materials for this study were collected from May 1986 to October 1989 in various parts of Estonia. The Rumpo Botanical Sanctuary on Vormsi Island (West Estonian Archipelago Biosphere Reserve, WEABR) and the Koljaku Reserve near Altja (Lahemaa National Park, LNP) were more thoroughly investigated. Lichen samples from the western coast of Estonia (Keibu, Lohusalu, Häädemeeste), Hiiumaa Island (WEABR), Põlva County (South Estonia), Harju County (North Estonia) were compared. Background content of radionuclides in lichens was determined by using herbarium samples from the collections of the Tallinn Botanical Garden, Estonian Academy of Sciences.

The Botanical Sanctuary on the Rumpo Peninsula lies on the southern part of Vormsi Island. The territory is covered with glacial, fluvioglacial and sea sediments. The peninsula itself is geologically young, formed only 3000—2000 years ago. The present configuration of the peninsula was formed mostly at the end of Phase V of the Limnean Sea. The main plant communities on the Rumpo Peninsula are natural meadow-type coastal associations (Laasimer, 1965): *Ditricho — Sedo — Thymetum*, *Medicagini — Festucetum rubrae* and *Clauco — Juncetum gerardii* with *Juniperus communis* stands. Light carbonaceous soils, sea sand and gravel predominate. On the Rumpo Peninsula such lichen species are found which belong to the Arctic flora element (Трасс, 1970), e. g. *Cetraria cucullata* and *C. nivalis* which are rare for Estonia but wide-spread in the Subarctic and Arctic.

The Koljaku Reserve lies in the northern part of the Lahemaa National Park. Vegetation of the reserve consists of lichen pine forests which grow on the Litorina Sea terrace. Soils are mostly carbonaceous. In other sample collection sites also dry lichen pine stands were chosen for determining the content of radionuclides in lichens.

In addition to the material collected systematically in Estonia also some lichen samples from North, Central and South Urals, the Baikal Nature Reserve, West-Caucasus and West-Spitsbergen were used for comparison. In this study we used mostly terricolous macrolichens *Cetraria cucullata* (Bellardi) Ach., *C. delisei* (Bory ex Schaerer) Nyl., *C. islandica* (L.) Ach., *C. nivalis* (L.) Ach., *Cladina arbuscula* (Wallr.) Hale et Culb., *Cl. mitis* (Sandst.) Hustich, *Cl. rangiferina* (L.) Nyl., *Cl. stellaris* (Opiz) Brodo, and the epiphytic *Hypogymnia physodes* (L.) Nyl.

Lichen samples on each site were collected by random selection method from a 20×20 cm plot. On the plots the cover degree of each species and biomass in air-dry state were determined. From the same plots the samples of litter (remains of dead plants) and the upper layer of soil (0—2 cm, in some cases 0—2—5 cm) were taken. The total number of samples analyzed was 156.

To determine the concentration of ^{137}Cs and ^{90}Sr in the plant material, it was cleaned and ashed, the soil was heated at 450—500 °C. The concentration of ^{90}Sr was determined by daughter isotope ^{90}Y, ^{137}Cs together with ^{134}Cs were determined by gamma-spectrometer of the type AI-256-6 (Нифонтова, Куликов, 1990). The statistical error did not exceed 10—15%.

Results and Discussion

According to the data by Molchanova and others (Молчанова et al., 1990) the maximum concentration of radionuclides in the soil and plants in the vicinity of the Chernobyl Nuclear Power Station amounted to 200 for ^{90}Sr, 500 for ^{134}Cs, and more than 2000 kBq/kg for ^{137}Cs. The isotope ratio $^{137}Cs/^{90}Sr$ amounted to 16—25 in leaf fallout and litter, and to 8—3 units in soil at the depth of 2 cm.

Table 1 shows background concentrations of ^{137}Cs and ^{90}Sr determined during 1971—1985 in three macrolichen species: *Cetraria islandica*, *Cladina rangiferina* and *Cl. stellaris*. The data show that the highest concentrations of these isotopes were found in the Urals where the concentrations 2.0 kBq/kg of ^{137}Cs and 0.6 kBq/kg of ^{90}Sr for *Cl. rangiferina* were relatively high in comparison with the other regions.

Table 1

Concentration of ^{137}Cs and ^{90}Sr (Bq/kg) in lichens *Cetraria islandica, Cladina rangiferina* and *C. stellaris* in different geographical locations, 1971—1985

Location	Lichen species	Concentrations	
		^{137}Cs	^{90}Sr
	Cetraria islandica		
Leningrad District	„ „	510	20
Estonia	„ „	140—190	3—30
	Cladina rangiferina		
Urals	„ „	300—1960	70—630
Kazakhstan	„ „	300	190
Baikal Nature Reserve	„ „	810	440
Estonia	„ „	60—150	4—30
	Cladina stellaris		
Urals	„ „	1080	160
Yamal Peninsula	„ „	1150	220
Estonia	„ „	160	10

Fig. 1 shows the maximum concentrations of ^{137}Cs and ^{90}Sr in lichens belonging to the *Cladina* genus in Estonia before (1) and after (2) the Chernobyl accident in comparison with the maximum concentrations of these radionuclides determined in lichens during the period of nuclear testing (1959—1965) in the Arctic (Троицкая et al., 1971). It can be seen that the concentrations of radiocaesium in lichens found in Estonia in 1986 exceeded those known from earlier publications two or even more times. The maximum concentration of radiostrontium reached the level known from literature.

In Fig. 2 there are two profiles — south-north (columns 1—3) from the Caucasus to Spitsbergen and west-east from Poland to Lake Baikal (columns 4—10) which reflect the maximum concentrations of ^{137}Cs and ^{90}Sr observed in lichens belonging to different genera or groups. In June 1986 relatively high values of the studied radionuclides concentrations were determined in epiphytic lichens in a mixed sample of *Usnea* and *Bryoria* sp. sp. from the West Caucasus (the Gangansky Range near Sochi), reaching 27.7 kBq/kg of ^{137}Cs and 3.0 kBq/kg of ^{90}Sr. Seaward and others (1988) measured the concentration of radiocaesium in epilithic lichen *Umbilicaria cylindrica* (L.) Delise ex Dubý in the Karkonosze

Mountains (Poland) in August 1986 and it amounted to 18.3 kBq/kg. A similar concentration of ^{137}Cs — 21.9 kBq/kg was measured in epiphytic lichen *Hypogymnia physodes* near the Beloyarskaya Nuclear Power Station in the Central Urals in August-September 1986. The highest concentration of the caesium radionuclide in lichens collected in Estonia in

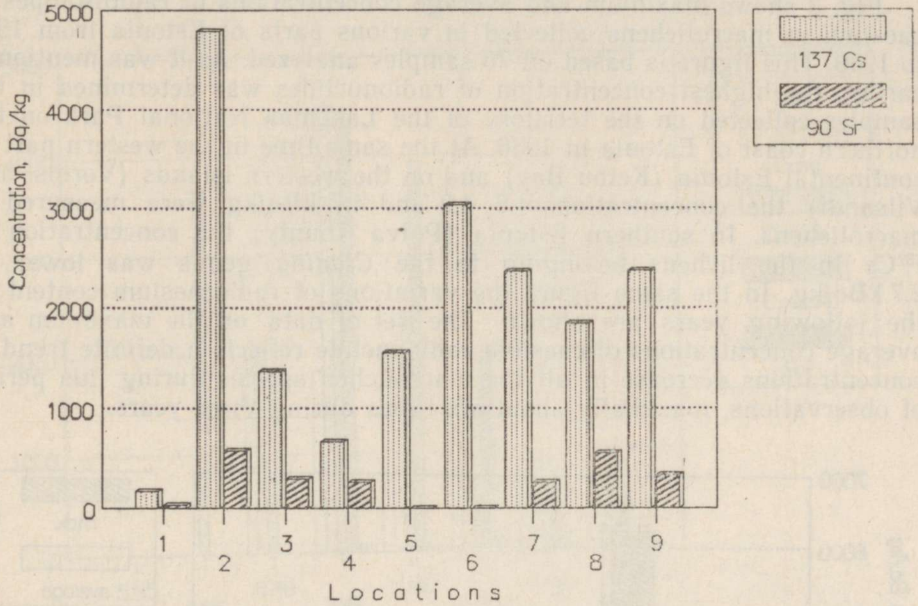

Fig. 1. Maximum concentrations of ^{137}Cs and ^{90}Sr in *Cladina* sp. sp. Estonia: 1 — 1985, 2 — 1986; 1959—1965: 3 — Alaska, 4 — Greenland, 5 — Norway, 6 — Sweden, 7 — Finland, 8 — Murmansk Region, USSR, 9 — Komi ASSR, USSR.

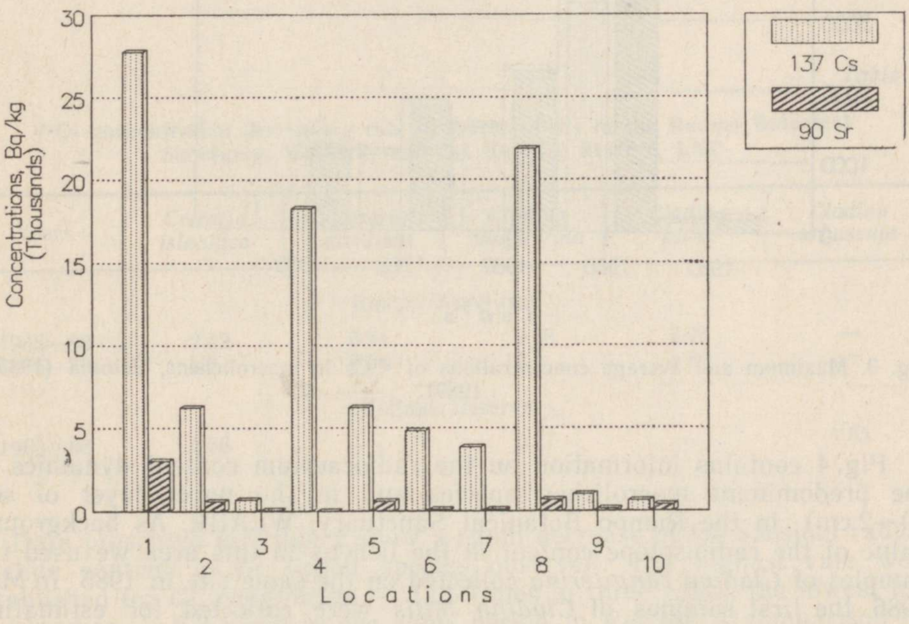

Fig. 2. Maximum concentrations of ^{137}Cs and ^{90}Sr in two profiles: I — south-north: 1 — Caucasus (Gangansky Range), 2 — Estonia, 3 — Spitsbergen; and II — west-east: 4 — Poland (Karkonosze Mountains), 5 — Estonia, 6 — Leningrad District, 7 — North Urals, 8 — Central Urals, 9 — South Urals, 10 — Baikal Nature Reserve.

1986 was found in the terricolous lichen *Cetraria islandica* in the Lahemaa National Park — 6.2 kBq/kg. It could be added that in the mushrooms (*Lactarius* sp.) picked in September 1986 in East-Virumaa (North-East Estonia) the concentration of radiocaesium was 16.6 kBq/kg in one sample, exceeding the maximum concentrations measured for Estonian mushrooms by Parmasto and Liiva (1988) three times.

Fig. 3 shows maximum and average concentrations of radioisotopes of caesium in macrolichens collected in various parts of Estonia from 1985 to 1989. This figure is based on 75 samples analyzed. As it was mentioned earlier, the highest concentration of radionuclides was determined in the samples collected on the territory of the Lahemaa National Park on the northern coast of Estonia in 1986. At the same time in the western part of continental Estonia (Keibu Bay) and on the western islands (Vormsi and Vilsandi) the concentrations 4.0, 4.9 and 4.7 kBq/kg were measured in macrolichens. In southern Estonia (Põlva County) the concentration of ^{137}Cs in the lichens belonging to the *Cladina* genus was lower — 2.7 kBq/kg. In the same figure the variations of radiocaesium content in the following years are shown. The set of data on the maximum and average concentrations of caesium radionuclide reflects a definite trend — concentrations decrease in all the macrolichen species during this period of observations, maximally about six times during three years.

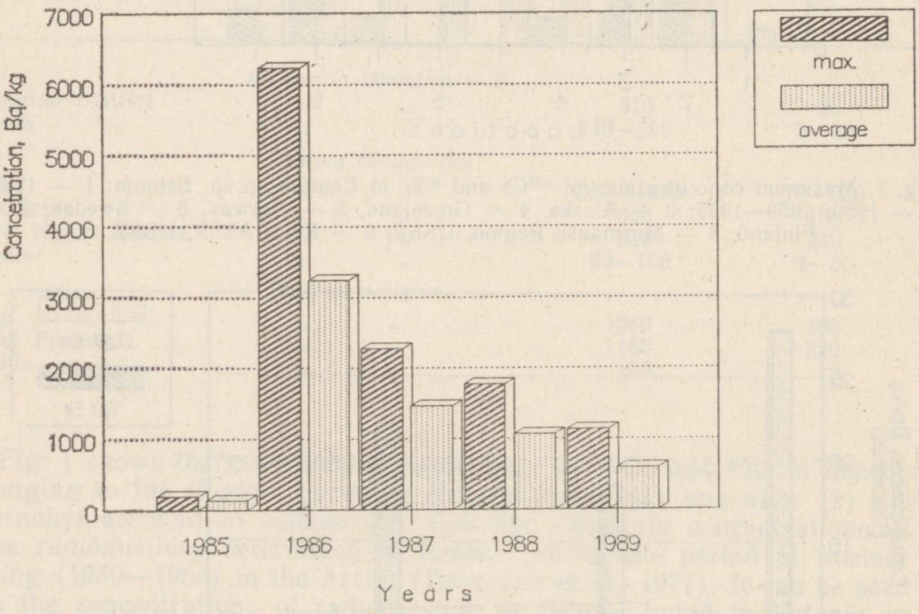

Fig. 3. Maximum and average concentrations of ^{137}Cs in macrolichens, Estonia (1985—1989).

Fig. 4 contains information on the radiocaesium content dynamics in the predominant macrolichen species and in the upper layer of soil (0—2 cm) in the Rumpo Botanical Sanctuary, WEABR. As background value of the radioisotope content in the lichens in this area we used the samples of *Cladina rangiferina* collected on the same site in 1985. In May 1986 the first samples of *Cladina mitis* were collected for estimating probable fallout of radionuclides after the Chernobyl accident. The concentration of ^{137}Cs in this sample was 3.7 kBq/kg — the highest for this lichen species for the whole observation period. From June the sampling was continued by using a randomized sample selection method. The same was done in July and November 1986 and in the following years. As it is seen from Fig. 4, the highest concentrations of radiocaesium were estimated in July 1986 — 4.5 for *Cl. rangiferina*, and 4.4 kBq/kg for *Cetraria cucullata*, except for *C. islandica* in which the maximum content, 2.1 kBq/kg, was measured in November 1986. In October 1987 the highest concentration of radiocaesium — 0.2 kBq/kg — was measured in sandy soil.

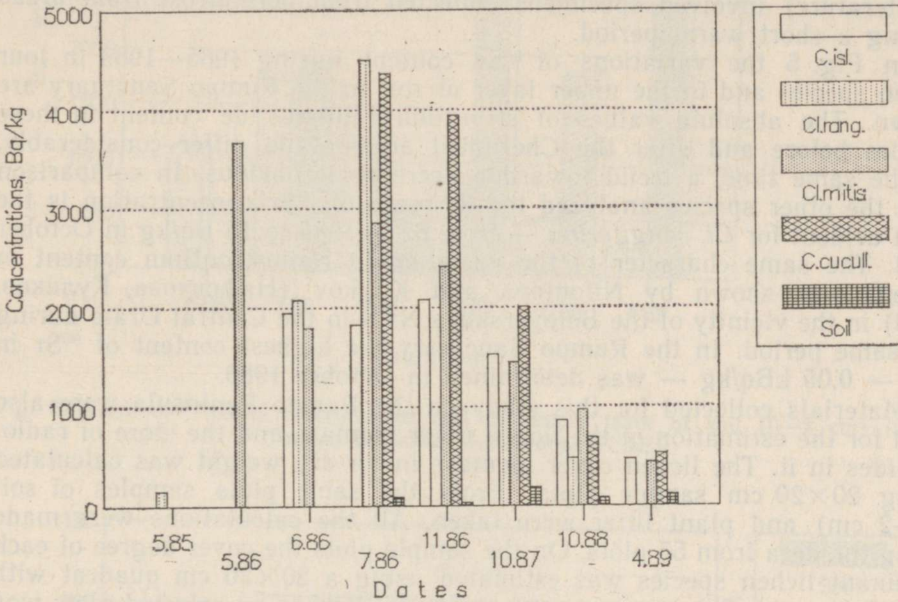

Fig. 4. Concentration of ^{137}Cs in macrolichens and in the upper layer of soil (0—2 cm) in the Rumpo Botanical Sanctuary, WEABR.

Table 2

^{137}Cs concentration decreasing rate in macrolichens in the Rumpo Botanical Sanctuary, WEABR, and the Koljaku Reserve, LNP

Years	*Cetraria islandica*	*Cetraria cucullata*	*Cladina rangiferina*	*Cladina mitis*	*Cladina arbuscula*
			Rumpo Sanctuary		
1986—88	2.42	6.31	9.38	3.79	—
1986—89	5.3	9.84	12.5	8.84	—
			Koljaku Reserve		
1986—88	5.66	—	2.37	—	1.53

Data presented in Table 2 show a rapid decrease of the caesium radionuclide content in all lichen species observed. The highest rate was established for *Cl. rangiferina* — 12.5 times in three years, the lowest for *C. islandica* — 5.3 times in the same period in Rumpo. A comparison of the decrease rates in Rumpo and Koljaku for two years reveals that for Koljaku the rate is lower for *Cl. rangiferina* — 2.4, but higher for *C. islandica* — 5.7.

It has been mentioned repeatedly in various publications that a twofold decrease of radionuclide concentration usually takes many years: 9—12 (Liden, Gustafsson, 1967), 3—13 (Martin, Koranda, 1971), 4—5 (Miettinen, Häsänen, 1967). The considerably high decrease rate of radionuclides in Estonian lichens could be explained by three circumstances: 1) the breaking up of ^{134}Cs, 2) areas selected for sampling sites are located in geologically young areas and have light carbonaceous soils with unstable ion balance, and 3) observations with Arctic lichens (mentioned in literature) involved specimens collected from permafrost from areas having a short warm period.

In Fig. 5 the variations of ^{90}Sr content during 1985—1989 in four lichen species and in the upper layer of soil in the Rumpo Sanctuary are shown. The absolute values of strontium radionuclide content in these species before and after the Chernobyl accident do differ considerably. At the same time, a trend toward a decrease is obvious. In comparison with the other species analyzed the decrease of ^{90}Sr concentration is the most evident for *Cl. rangiferina* — from 62 in 1986 to 15 Bq/kg in October 1988. The same character of the variation in radiostrontium content in lichens was shown by Nifontova and Kulikov (Нифонтова, Куликов, 1990) in the vicinity of the Beloyarskaya NPS in the Central Urals during the same period. In the Rumpo Sanctuary the highest content of ^{90}Sr in soil — 0.05 kBq/kg — was determined in October 1988.

Materials collected for this study on the Rumpo Peninsula were also used for the estimation of the lichen cover biomass and the store of radionuclides in it. The lichen cover biomass in air-dry weight was calculated using 20×20 cm sample plots. From the same plots samples of soil (0—2 cm) and plant litter were taken. All the calculations were made using the data from 55 plots. On the sample plots the cover degree of each dominant lichen species was estimated using a 20×20 cm quadrat with a 2×2 cm grid. The average cover degree of lichens on selected plots was 84.5% and average biomass 0.58 kg/m². In Koljaku these figures were as follows: 15 analyzed plots, average cover degree — 81.7%, and average biomass — 0.62 kg/m².

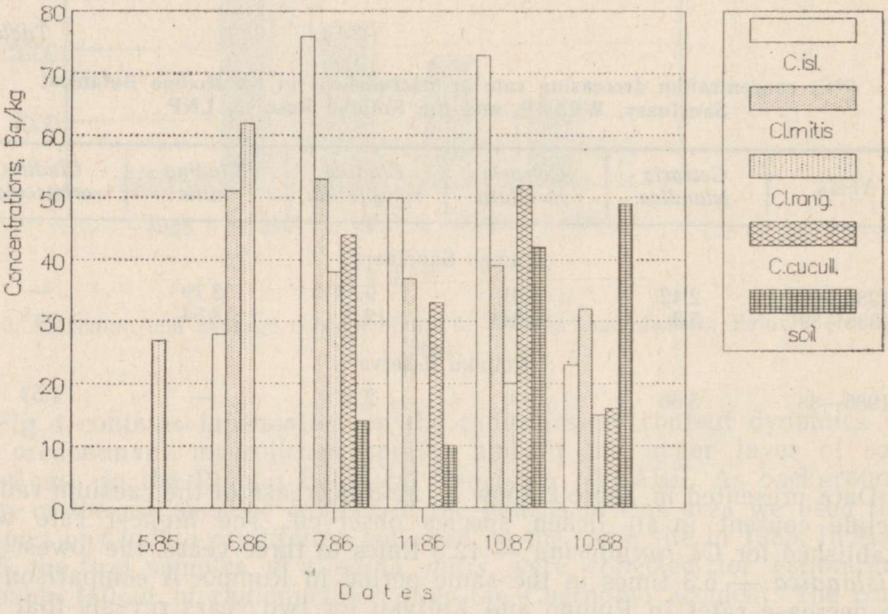

Fig. 5. Concentration of ^{90}Sr in macrolichens and in the upper layer of soil (0—2 cm) in the Rumpo Botanical Sanctuary, WEABR.

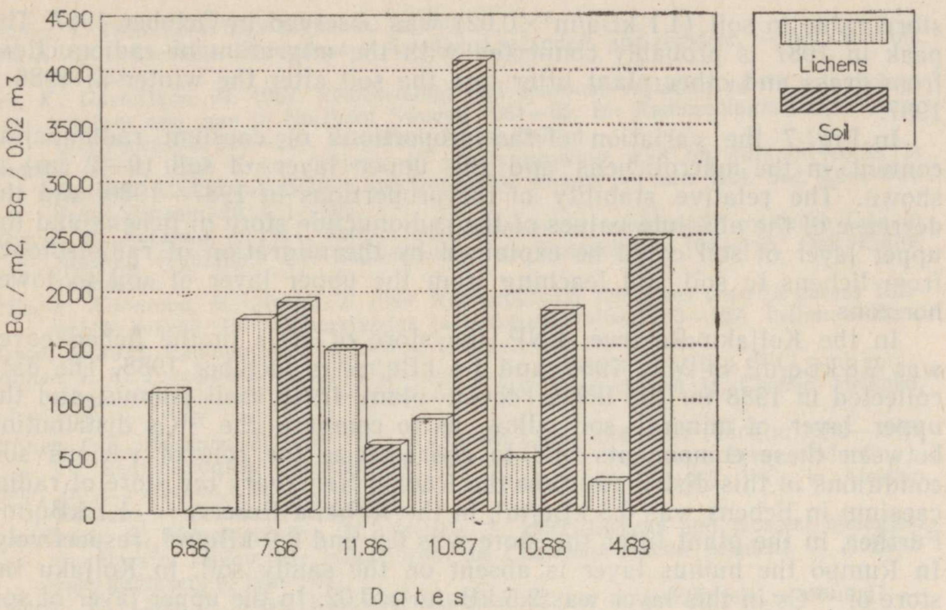

Fig. 6. Store of ^{137}Cs in macrolichens and the upper layer of soil (0—2 cm) in the Rumpo Botanical Sanctuary, WEABR.

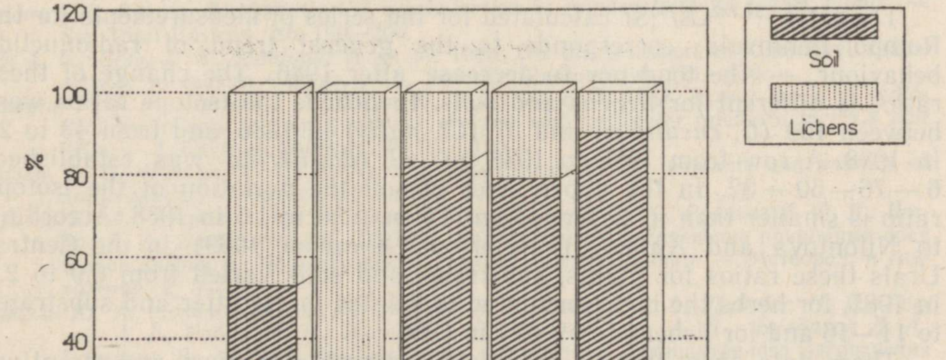

Fig. 7. Variation of the proportions of ^{137}Cs store in macrolichens and in the upper layer of soil (0—2 cm) in the Rumpo Botanical Sanctuary, WEABR.

Fig. 6 shows the variation of the radionuclide store in the lichen cover on the Rumpo Peninsula during 1986—1989. The same trend as in the case of radiocaesium concentration variations is evident — in July 1986 the store of ^{137}Cs in the lichen cover was the highest, 1.7 kBq/m^2, and then it decreased continuously reaching 0.23 kBq/m^2 in 1989. The character of the variation in radiocaesium store in the soils is different. The highest store value in soil (4.1 kBq/m^3×0.02) was observed in October 1987. The peak in 1987 is probably connected with the migration of radionuclides from grass and other plant litter into the soil after the winter of 1986—1987.

In Fig. 7 the variation of the proportions of caesium radionuclide content in the macrolichens and the upper layer of soil (0—2 cm) is shown. The relative stability of the proportions in 1987—1989 and the decrease of the absolute values of the radionuclide store in lichens and the upper layer of soil could be explained by the migration of radioisotopes from lichens to soil and leaching from the upper layer of soil to lower horizons.

In the Koljaku Reserve, LNP, the store of ^{137}Cs in the lichen cover was 2.8 kBq/m^2 in May 1986, and 1.1 kBq/m^2 in October 1988. The data collected in 1988 for the lichen cover, plant litter, soil humus, and the upper layer of mineral soil allow us to compare the ^{137}Cs distribution between these components and to characterize the role of various soil conditions in this distribution: in the Rumpo Sanctuary the store of radiocaesium in lichens was 0.5 kBq/m^2, in the Koljaku Reserve — 1.1 kBq/m^2. Further, in the plant litter the store was 0.2 and 0.9 kBq/m^2, respectively. In Rumpo the humus layer is absent on the sandy soil, in Koljaku the store of ^{137}Cs in this layer was 2.5 kBq/m^3×0.02. In the upper layer of soil (0—2 cm) the store was 1.9 in Rumpo and 1.1 kBq/m^3×0.02 in Koljaku. These data show that in the case of young mineral soils the migration of radiocaesium from lichens to soil and deeper into the soil is more intensive. In developed soils the humus layer is the depot of radiocaesium and further migration deeper into the soil profile is decelerated.

The ratio ^{134,137}Cs/^{90}Sr calculated for the series of measurements for the Rumpo Peninsula corresponds to the general trend of radionuclide behaviour — the tendency to decrease after 1986. The change of these ratios is different for lichens and soils. In lichens the isotope ratios were between 109 (*C. cucullata*) and 37 (*Cl. mitis*) in 1986, and from 43 to 23 in 1988. A row from 1985 to 1988 for *Cl. rangiferina* was established: 6 — 76— 50 — 32. In the upper layer of soil the variation of the isotope ratio is smaller than in lichens: from 4.5 in 1986 to 1.5 in 1988. According to Nifontova and Kulikov (Нифонтова, Куликов, 1989) in the Central Urals these ratios for plants, substrates and soils varied from 0.9 to 2.2 in 1985, for herbs the ratio amounted to 6.4, for forest litter and substrates to 11—16 and for lichens to 19—74 in 1986.

These data show that in Estonia a noticeable caesium and strontium radionuclide pollution still exists caused by the Chernobyl accident in 1986, and that the character of the isotope behaviour is conditioned by site conditions. Some differences in the radioisotope content in macrolichens collected from different parts of Estonia serve as an evidence of pattern-like fallout of these isotopes. And finally, the character of Cs/Sr ratio confirms the predominant radiocaesium output into the atmosphere and its long-range transportation during the Chernobyl accident.

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