Radionuclide distribution in components of the Sarbalyk limnetic system (Baraba lowland, Western Siberia)

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Geochemical and mineralogical analysis of the bottom sediment cores of small lakes can be used as the basis for radio-ecological monitoring and environmental protection measures. Here, features of the radionuclide activity distribution in components of the limnetic ecosystems of the central Baraba lowland were studied, and factors affecting radiocesium absence in the bottom sediment of Lake Sarbalyk were identified. Fieldwork included the sampling of limnetic ecosystem components, bottom sediment weighing, and the determination of pH, Eh, TDS and oxygen content. Subsequent laboratory investigation involved the determination of major and trace element concentrations by atomic absorption spectrometry and X-ray fluorescence analysis; mineral composition by X-ray diffractometry (XRD); analysis of sample morphology and composition via scanning electron microscopy; and natural radionuclide and radiocesium content determination by the gamma-spectrometric method. Radiocesium is present in all the studied lake bottom sediments of the central Baraba lowland in the upper horizons, with the exception of Lake Sarbalyk. Limnetic ecosystem components were analyzed using modern analytical methods, considering the distribution features of natural and artificial radionuclides in catchment area soils, tussocks and the bottom sediment of Lake Sarbalyk. The results revealed that the absence of radiocesium in the bottom sediment of Lake Sarbalyk is due to two factors: the characteristics of the lake overgrowth and the sorption of dissolved forms of ^{137}Cs. *KEYWORDS:* Radiogenic isotope geochemistry; small lake; Western Siberia.

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Introduction

Small lakes are an important object of study for experts in various modern scientific fields. Bottom sediment (BS) is one of the main components of lacustrine ecosystems and its genesis interconnects with processes occurring in both the catchment area and the lake itself. An important feature of BS is that any change, either directly in the water system or in its environment, will be "documented" by depth [*Lan et al.,* 2018; *Zhang et al.,* 2018]. Since small lakes are more sensitive to changes than large water bodies, their study can provide answers to many scientific questions

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concerning natural and anthropogenic impacts on the environment, occurring in the past, present or future [*Bihari et al.,* 2018; *Egorov et al.,* 2018; *Pestryakova,* 2016; *Solotchina,* 2009; *Subetto,* 2016; *Wang et al.,* 2018].

Southwestern Siberia contains more than 20 thousand lakes of different sizes, water regimes and salinities. Most are inland, shallow, fresh to salt, and form the major component of the hydrography of the Ob-Irtysh interfluve [*Bejrom et al.,* 1986; *Shtin,* 2005]. The small lakes are characterised by active sapropel formation, which in some bodies is progressive with a constant increase in sediment volume against terrigenous material transport and eolian processes, mainly in humid environments [*Naumova et al.,* 2013; *Roslyakov et al.,* 2013].

Sapropel is a bottom sediment of water bodies that is formed as a result of biochemical, microbiological and mechanical processes from the remains of dying plants and animals, which introduce organic and mineral impurities. Sapropel consists of four components: mineral, organic, bioactive and aquatic [*Strakhovenko et al.,* 2016]. Such variation in both genesis and composition results in many different types of sapropel with varying characteristics [*Korde,* 1969; *Lopotko,* 1978; *Sukachyov,* 1943]. The most dominant factors include the composition of mineral particles and organic matter, as well as their ratio. Several features characterise typical sapropelic sediments: a tobacco-green colour that varies to black under reduction conditions, a colloidal structure, and occasionally the smell of hydrogen sulfide. Sapropels can also be classified based on their relative organic and mineral composition (ash content) into four main types: *organogenic* with ash content up to 30%; *organomineral* (30–50%); *mineral-organogenic* (50–70%); and *mineralised* (70–85%). BS with ash content above 85% are defined as mineral silts. The chemical composition of the mineral component may vary, although SiO_2 and CaO strongly dominate, with the Si/Ca ratio also used to allocate classes of sapropel: *silicon* (Si > Ca); *calcium* (Ca > Si); and *mixed* (Si $\sim$ Ca) [*Strakhovenko et al.,* 2016].

The lakes selected for analysis in the present study are located within the central part of the Baraba lowland in Western Siberia (Figure 1). The study region is typical of Central Baraba: In the southwest, the land surface is marked by strictly parallel, north-eastern strike, alternating hills and hollows, with numerous small and large lakes, marshes and areas of dry land. Because of this landscape there is local redistribution of moisture and easily soluble salts, flowing from hills to hollows [*Il'in and Syso,* 2001; *Rikhvanov,* 2009].

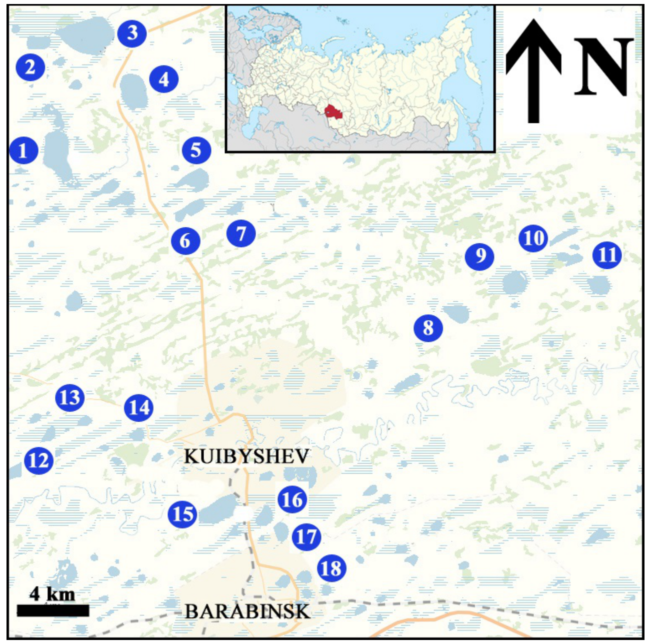

Figure 1. Location scheme of the studied lakes: 1 – Bol. Kazatovo, 2 – Chylym, 3 – Barchin, 4 – Kambala, 5 – Kaily, 6 – Bergul, 7 – Yargol, 8 – Suetok, 9 – Bilgen, 10 – Bol. Kurgan, 11 – Sarbalyk, 12 – Tsybovo, 13 – Zhiloe, 14 – Mostovoe, 15 – Bol. Kaily, 16 – Peschanoe, 17 – Chistoe, 18 – Bugristoe.

Natural background radiation is determined by the content of natural radioactive elements including uranium, thorium and potassium. The central part of the Western Siberian plate belongs to a zone of low radiation, because the natural radionuclides contained in the region's rocks and soils present low radiation dose values. However, since these elements are found in all natural objects in certain quantities, and their increased concentrations can be potentially dangerous, it is essential that sapropels are analysed regarding their ecological safety, in terms of their content of toxic elements, as well as natural and artificial radioactivity [*Rikhvanov,* 2009].

Southwestern Siberia has been contaminated by technogenic radionuclides since the beginning of nuclear weapons testing at the Semipalatinsk and Novaya Zemlya nuclear test sites in 1949. The distribution of ^{137}Cs pollution in the study area is mosaic in form, and varies in cause and scale, depending particularly on the preservation degree of primary mosaic contamination under the influence of secondary redistribution. The primary mosaic radiation distribution is itself determined by nuclear test types, atmospheric conditions, the radionuclide fractionation effect during transfer, and the nature of radioactive cloud movement [*Strakhovenko et al.,* 2010]. Soil ^{137}Cs distribution heterogeneity was established by comparing samples taken in the same elementary landscape type at sites equidistant from each other and without significant differences in relief and sod layer. Radiocesium redistribution in soil after deposition depends on many specific factors, including soil type, rainfall, drainage, landscape and vegetation. Statistical estimates are thus more robust than single estimates, and can be reproduced satisfactorily at a significance level of 0.05 and with a sufficient sample size. Data can also be distributed over entire catchment areas, producing a generalised characterisation of current radiation pollution.

A large group of scientists have carried out work estimating the global content of ^{137}Cs in the total contamination of Western Siberia soils, with a value of 43 mCi/km^{-2} determined in 2000. For most studied areas, total pollution levels of ^{137}Cs in Siberian lacustrine BS and in the soils of their catchment area exceed the global radioactive background level. Radiocesium distribution heterogeneity in BS is influenced by many factors, primarily the unevenness of precipitation during nuclear tests in reaching lake waters and catchment area soils. Furthermore, the configuration of primary habitats may vary under the influence of erosion-accumulative processes and lithochemical migration. In the study area, contamination of lacustrine BS is highlighted sharply against the background, especially in Lake Sarbalyk. Indeed, an initial test in 1995 revealed that Lake Sarbalyk BS radiocesium activity concentration was equal to zero, with re-samplings in 2012 and 2014 confirming this first result [*Ovdina et al.,* 2016].

The aim of the present work was thus to study the features of radionuclide activity concentration distribution in the lacustrine ecosystems of the central Baraba lowland and to identify the factors affecting the radiocesium absence in Lake Sarbalyk bottom sediment.

Materials and Methods

Materials

Bottom sediments. Stratified sediment cores were sampled in all the studied lakes, the bottom sediments of which were identified as sapropels. Bottom sediment sampling was carried out according to standard methods. BS were collected from a catamaran using a cylindrical sampler with a vacuum shutter (diameter 82 mm, length 110 cm) developed by the Taifun Research and Production Association, Russia. BS cores were sampled at an interval of 3–5 cm to a depth of 50–120 cm; each sample was numbered in order and packed in a plastic bag.

Soils. Soil samples were taken using a metal ring sampler [*Malikova et al.,* 2005] with a diameter of 82 mm and height of 50 mm, through the depth of the entire soil section. All soil samples were dried and weighed. Soil was also sampled from the catchment area of Lake Sarbalyk, beginning on a hill and ending on the lakeshore. Soil types followed the sequence: leached chernozem$\longrightarrow$ grey forest soil$\longrightarrow$ saline meadow carbonate soil$\longrightarrow$ meadow-marsh carbonate soil.

Water. Water sampling for all analyses was carried out according to standard methods. For this purpose, bottles of volume 1, 0.5 (both plastic, to determine major and trace element compositions) and 0.33 l (glass, to determine Hg content) were used, with water for trace element composition and Hg tests preserved using nitric acid.

Tussocks. Tussocks were divided into upper and lower parts, both of which were sampled: upper part – living reed and soil particles from the lake catchment area; lower part – tightly intertwined roots.

Biota. Primary sapropel-forming material (phytoplankton, photosynthetic pigments, zooplankton, phytoperiphyton and phytobenthos, macrophytes) was sampled for genesis determination at each lake [*Zarubina,* 2013].

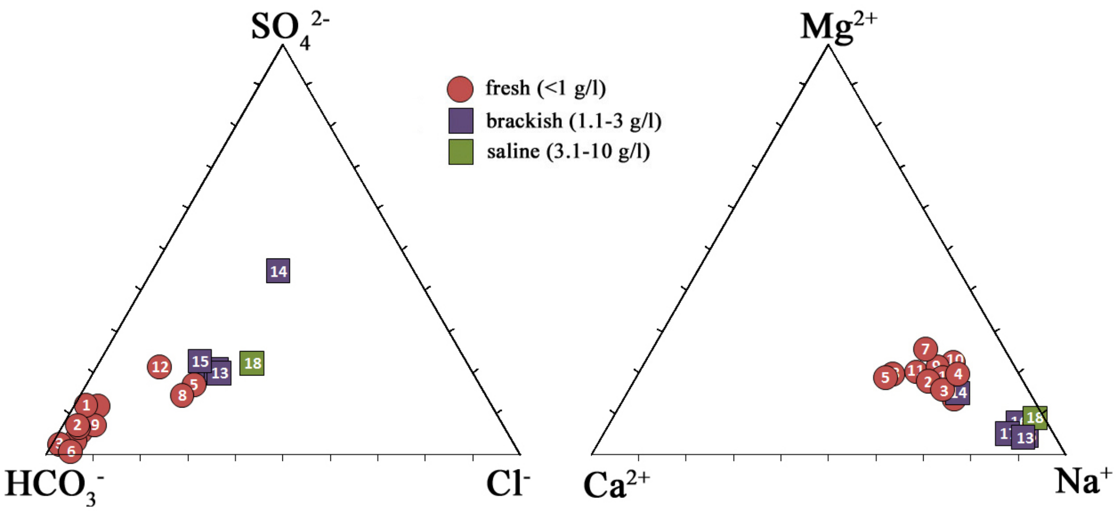

Figure 2. Ferre triangle of lake water composition (%-eq) with respect to TDS values. Lakes: 1 – Suetok, 2 – Bilgen, 3 – Sarbalyk, 4 – Mostovoe, 5 – Kazatovo, 6 – Yargol, 7 – Bergul, 8 – Kambala, 9 – Chylym, 10 – Bol. Kurgan, 11 – Barchin, 12 – Kaily, 13 – Bol. Kaily, 14 – Tsybovo, 15 – Chistoe, 16 – Zhiloe, 17 – Bugristoe, 18 – Peschanoe.

Methods

Physical and chemical variables of water and BS material were recorded in situ (pH, Eh, TDS) using an ANION-7000 portable liquid analyser (Biomer, Russia). Measurement of O_2 content was carried out via the biochemical oxygen consumption method for five days (BOD5), enabling the establishment of the extent of water pollution based on the amount of oxygen consumed after the aerobic biochemical decomposition of organic substances in the test water (at 20°C, in mg^{-1}). Determination of BOD5 was carried out in the initial sample based on the difference in oxygen content before and after incubation for five days without access to oxygen and light. Chemical composition analyses of BS samples were carried out at the Analytical Center for multi-elemental and isotope research SB RAS, and at the Laboratory of Geochemistry of Noble and Rare Elements of IGM SB RAS, Novosibirsk. BS and water major and trace element compositions were determined via atomic absorption using a Solaar M6 instrument (Thermo Electron, UK) equipped with a Zeeman and deuterium background corrector. Major element composition was determined by X-ray fluorescence analysis (ARL-9900-XP, Applied Research Laboratories, USA). X-ray diffractometry (XRD) was used to determine sample mineral composition (ARLX'TRA, Thermo Fisher Scientific (Ecublens) SARL, Switzerland). Sample morphology, phase and chemical composition was determined using a scanning electron microscope (TESCAN MIRA3, Tescan, Czech Republic) equipped with an energy spectrometer (OXFORD, Oxford Instruments, UK). Determination of natural radionuclides and radiocesium was carried out via a gamma-spectrometric method using a well coaxial detector from ultrapure germanium (HPGe) with preamplifier and low-background cryostat (EGPC 192-P21/SHF 00-30A-CLF-FA; EURYSIS MESURES, France).

Results

We sampled a total of 27 lacustrine bottom sediment cores (413 samples), 15 soil cores (60 samples) and 78 water samples.

According to the obtained analytical data (main water ion content), water in the Baraba lakes is mainly bicarbonate magnesium-sodium or bicarbonate sodium, alkaline (pH 8.1–10) and fresh (0.2–0.6 g L^{-1}) (Figure 2), with occasional brackish (TDS 1–3 g L^{-1}) and salty (TDS 3.3 g L^{-1}) lakes. The water composition of Lake Sarbalyk is alkaline, sodium bicarbonate freshwater (TDS 0.3 g L^{-1}).

Field and analytical studies revealed that the BS of all the studied small lakes are sapropels, most of

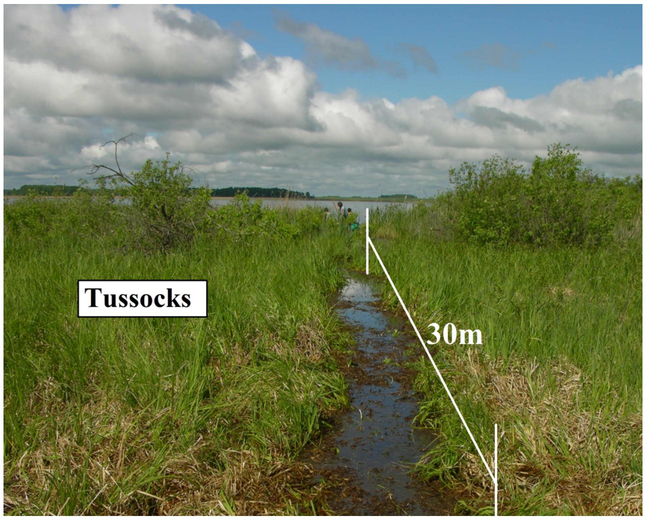

Figure 3. Lake Sarbalyk massive-thicket type of vegetation. Tussocks.

which are organic-mineral or mineral-organogenic, silicon in class and plankton-macrophyte in type. Lake Sarbalyk bottom sediment is organic-mineral silicon sapropel, and macrophyte in type based on prevailing productivity.

The contribution of different ecosystem biotic components to lacustrine bottom sediment formation can vary significantly, thereby affecting the sediment's final chemical and mechanical composition.

The main primary producers of organic matter in Lake Sarbalyk are higher aquatic plants – macrophytes. According to overgrowth degree and volume of primary productivity, Lake Sarbalyk belongs to a class of lakes with massive-thicket type of vegetation (Figure 3). Rigid air-water vegetation is represented by the communities of *Typha latifolia* and *Phragmites australis (Cav.) Trin. ex Steud.* that dominate the coastal zone in shallow water. Soft submerged vegetation communities of *Myriophyllum sibiricum Kom., Stratiotes aloides L.* and *Ceratophyllum demersum L.* are widespread in the main water area. The lake overgrowth area is about 70% [*Zarubina,* 2013]. During the vegetation period, macrophytes in Lake Sarbalyk form 1830.9 g m^{-2} per year of organic matter, including 1014.3 gC_{org} m^{-2}, 44.5 gN m^{-2} and 4.0 gP m^{-2}. Lake Sarbalyk is associated with minor phytoplankton contribution to primary product formation. The gross primary production value of phytoplankton during the study period was low and amounted to only 0.37 mgO_2 L^{-1} per hour, with 665 thousand cell dm^{-3} and biomass of 0.02 g m^{-3} [*Yermolaeva et al.,* 2016]. Estimated values of phytoplankton contribution to Lake Sarbalyk bottom sediments varied at 0.18 gC_{org} m^{-2} per year, 0.03 gN m^{-2} per year and 0.004 gP m^{-2} per year.

Zooplankton is actively involved in the processes of sedimentation, filtering phytoplankton and bacterioplankton and transforming it into fecal pellets. In Lake Sarbalyk zooplankton excreted up to 1.71 gP m^{-2} , up to 17.57 gNorg m^{-2}, up to 145.52 gCorg m^{-2} per year and due to the withering away of zooplankton in the lake bottom sediments comes up to 1137.86 mgCorg m^{-2}, 264.31 mgNorg m^{-2} and 3.97 mgP m^{-2} per year.

Almost the entire flow of matter, formed during the day in the upper layers of the lake, has time to reach the lake bottom without being fully mineralized in the water column due to the lake's shallow depth. Further transformation of both faecal and dying biological material occurs at the lake bottom as a result of the activity of bacteria and benthic organisms.

Thus, hydrobionts are the main source of autochthonous organic matter in the bottom sediments, largely determining the latter's chemical composition. Macrophytes and zooplankton make the main contribution to organic matter formation in Lake Sarbalyk.

Discussion

Analysis of Lake Sarbalyk bottom sediment composition did not reveal any anomalies in major and trace element distributions or mineral phases and, in general, sediment composition does not change with depth.

The BS composition of all the studied lakes was analysed in terms of natural and artificial radionuclide content (Table 1). According to the data obtained, the total specific activity (Ac) of natural radionuclides across all the lakes varies at around 11–84 Bq kg^{-1}, which corresponds to the radiation-hygienic standard for sapropels.

Radiocesium was not detected in Lake Sarbalyk bottom sediment, in contrast to nearby lakes (Lake Bol. Kurgan, Lake Bilgen and Lake Suetok) where radiocesium activity concentrations in bottom sediments range from 47–178 Bq kg^{-1}.

Table 1. Natural and Artificial Radionuclide Activity Concentrations in Small-Lake Bottom Sediments of the Central Baraba Lowland

№	Lake	^{232}Th, Bq kg^{-1}	^{238}U(Ra), Bq kg^{-1}	^{40}K, Bq kg^{-1}	Ac, Bq kg^{-1}	^{137}Cs, Bq kg^{-1}
1	Suetok	18.9	28.6	396.4	58	178
2	Bilgen	21.6	24.3	431.8	65	47
3	Sarbalyk	24.6	33.1	315.1	59	0
4	Mostovoe	23.1	19.0	250.5	52	12
5	Kazatovo	14.3	21.2	294.4	44	183
6	Yargol	16.1	49.8	151.3	34	24
7	Bergul	13.5	32.4	235.5	38	132
8	Kambala	22.0	25.4	252.9	50	78
9	Chylym	13.3	30.4	132.5	29	140
10	Bol. Kurgan	6.7	21.2	170.7	23	114
11	Barchin	4.2	43.1	65.8	11	209
12	Kaily	13.0	31.5	158.3	30	189
13	Bol. Kaily	28.7	26.0	359.3	68	96
14	Tsybovo	26.7	16.4	292.7	60	168
15	Chistoe	34.7	36.0	455.4	84	151
16	Zhiloe	23.1	30.3	424.2	66	115
17	Bugristoe	17.3	24.5	238.1	43	171
18	Peschanoe	19.0	28.1	112.8	35	75

Table 2. Natural and Artificial Radionuclide Activity Concentrations in Lake Sarbalyk Bottom Sediments in 1995, 2012 and 2014

Date of sampling	Sample number	Depth, cm	^{238}U(Ra), Bq kg^{-1}	^{232}Th, Bq kg^{-1}	^{40}K, Bq kg^{-1}	^{137}Cs, Bq kg^{-1}
September, 1995	Ku22Do1	0–10	2.80	6.00	0.51	0
	Ku22Do2	10–15	1.80	4.90	0.97	0
	Ku22Do3	15–25	2.30	5.30	0.35	0
June, 2012	B121Do1	0–5	5.0	8.4	1.26	0
	B121Do2	5–10	3.3	5.6	1.22	0
	B121Do3	10–15	2.5	6.1	1.09	0
	B121Do4	15–20	0.7		0.30	0
	B121Do5	20–25	1.9	6.9	1.09	0
	B121Do6	25–30	2.8	6.7	0.74	0
	B121Do7	30–35	2.1	4.8	0.77	0
	B121Do8	35–40	1.8	3.7	0.82	0
	B121Do9	40–45	2.0	6.2	0.90	0
	B121Do10	45–50	1.3	5.3	1.05	0
	B121Do11	50–55	2.7	6.0	1.10	0
	B121Do12	55–60	3.7	6.7	1.04	0
August, 2014	B145Do1	0–10	1.05	5.60	1.10	0

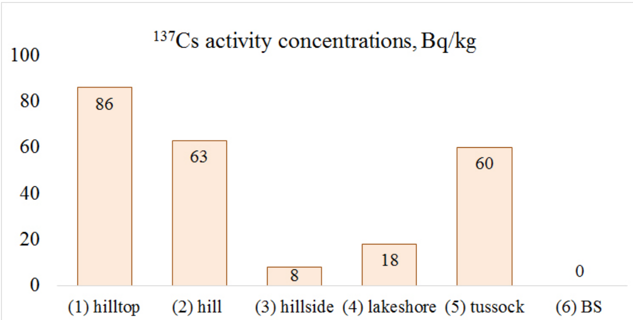

Figure 4. Radionuclide distribution in components of the Sarbalyk lacustrine ecosystem: 1 – leached chernozem, 2 – grey forest soil, 3 – saline meadow carbonate soil, 4 – meadow-marsh carbonate soil, 5 – tussock, 6 – lacustrine bottom sediments.

Lake Sarbalyk bottom sediments were sampled three times in 1995, 2012 and 2014. ^{137}Cs was absent in all years in bottom sediments samples of Lake Sarbalyk (Table 2).

Soils in the study catchment were also analysed in terms of their basic physical and chemical properties (granulometric composition, humus content, pH, radionuclide activity concentrations (^{238}U, ^{232}Th, ^{40}K, ^{137}Cs), major and trace element composition).

With respect to soil chemical properties, significant ^{137}Cs activity concentrations were determined only in the first 0–15 cm of soil cover in the Lake Sarbalyk catchment, with no radiocesium identified at lower depths. Radiocesium activity exceeded the global background by at least 2 times in cores of the leached chernozem (86 Bq kg^{-1}) and grey forest soils (67 Bq kg^{-1}) on the catchment hill. In contrast, concentrations in the saline meadow carbonate soil on the slope were only 8 Bq kg^{-1}, and in the meadow-marsh carbonate soil on the lakeshore 18 Bq kg^{-1}. These values likely reflect the high degree of turf cover contributing to radiocesium retention in the upper layer, preventing the eolian transport of soil particles.

In terms of tussock material, ^{137}Cs activity concentrations in these samples were much higher than those found in the saline meadow carbonate soil and meadow-marsh carbonate soil.

In summary, the distribution of radiocesium activity concentrations in components of the Lake Sarbalyk ecosystem has a saltatory character (Figure 4), with the highest levels found in soil cover on the hill (leached chernozem) and in tussocks, and an absence of radiocesium in lake bottom sediment. This pattern likely reflects the action of tussocks trapping soil particles containing radiocesium due to their tightly intertwined roots.

Conclusion

Analysis of the natural and artificial radionuclide distribution in soils, tussocks and bottom sediment of the Lake Sarbalyk catchment area revealed that the absence of radiocesium in the lake bottom sediments is due to several factors. In particular, the high degree of slope vegetation cover does not allow ^{137}Cs to enter the lake with eolian soil particles, while direct deposition of radiocesium onto the lake area at the time of nuclear tests did not occur.

1. Features of overgrowth. The lake overgrowth area comprises around 70% dense reeds, the stems, leaves and roots of which trap radiocesium-containing soil particles and prevent their transfer from the slope. Based on the mosaic pattern of radiocesium contamination on the surface, soils on the hill and slope are characterised by high activity concentrations. Below the slope, due to the weathering of soil particles, radiocesium levels are very low. Tussocks trap soil particles and prevent the deposition of radiocesium in the bottom sediment of Lake Sarbalyk.

2. The results indicate sorption of the dissolved forms of radiocesium and their accumulation in tussocks. Dissolved forms of radiocesium accumulate in the roots and lower parts of reeds, which together with soil particles from the slope increases radiocesium activity concentrations in these features.

Considering the unique distribution of radionuclides by components of the Sarbalyk lacustrine system, the data obtained in the present study can be used as the basis for radio-environmental monitoring and environmental protection measures.

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