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ОРИГИНАЛЬНАЯ СТАТЬЯ ORIGINAL ARTICLE

Fractions of trace metals in the sediments of permafrost-affected lakes in Northern Siberia, Lena delta

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Summary

The aim of the study is to estimate the role of sediment components in the accumulation of microelements in lakes of the permafrost-affected area (the Lena Delta, northern Siberia). A fractional analysis of several trace elements (V, Cr, Co, Ni, Cu, Zn, Sn, and Pb) was conducted. Samples were collected from 10 lakes during the summer period of the "LENA 2019" expedition. The content of the chemical elements in the sediments was measured with the ICP-MS instrument. The results of the work show a relatively homogeneous distribution of the acid-soluble forms of the metals in sediments from different lakes. The content of trace elements in all the lakes studied is determined by natural (lithogenic) sources. Stable minerals inherited from the rocks play a crucial role in the sediment formation. Most of the elements are predominantly fixed in stable mineral and organometallic fractions. However, for V, Co, and Cr a high level of geochemical mobility was identified in some of the lakes. The metals of soluble complexes are capable of migrating from the sediments to the water due to physical and chemical changes in the aquatic environment. Furthermore, the organic substances and the Fe/Mn hydroxides of the sediments have a low potential ability to bind the metals into stable compounds.

Keywords: fractions of metals, lake sediments, Lena Delta, permafrost, Siberia.

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INTRODUCTION

The Lena Delta is located in a permafrost-affected area of the Russian Arctic (Fig. 1). There are numerous lakes within the territory. Their hydrological and hydrochemical features are predominantly affected by surface fluxes from the catchment areas, the groundwater of the active layer, and the flooding regime of the river Lena [1–3].

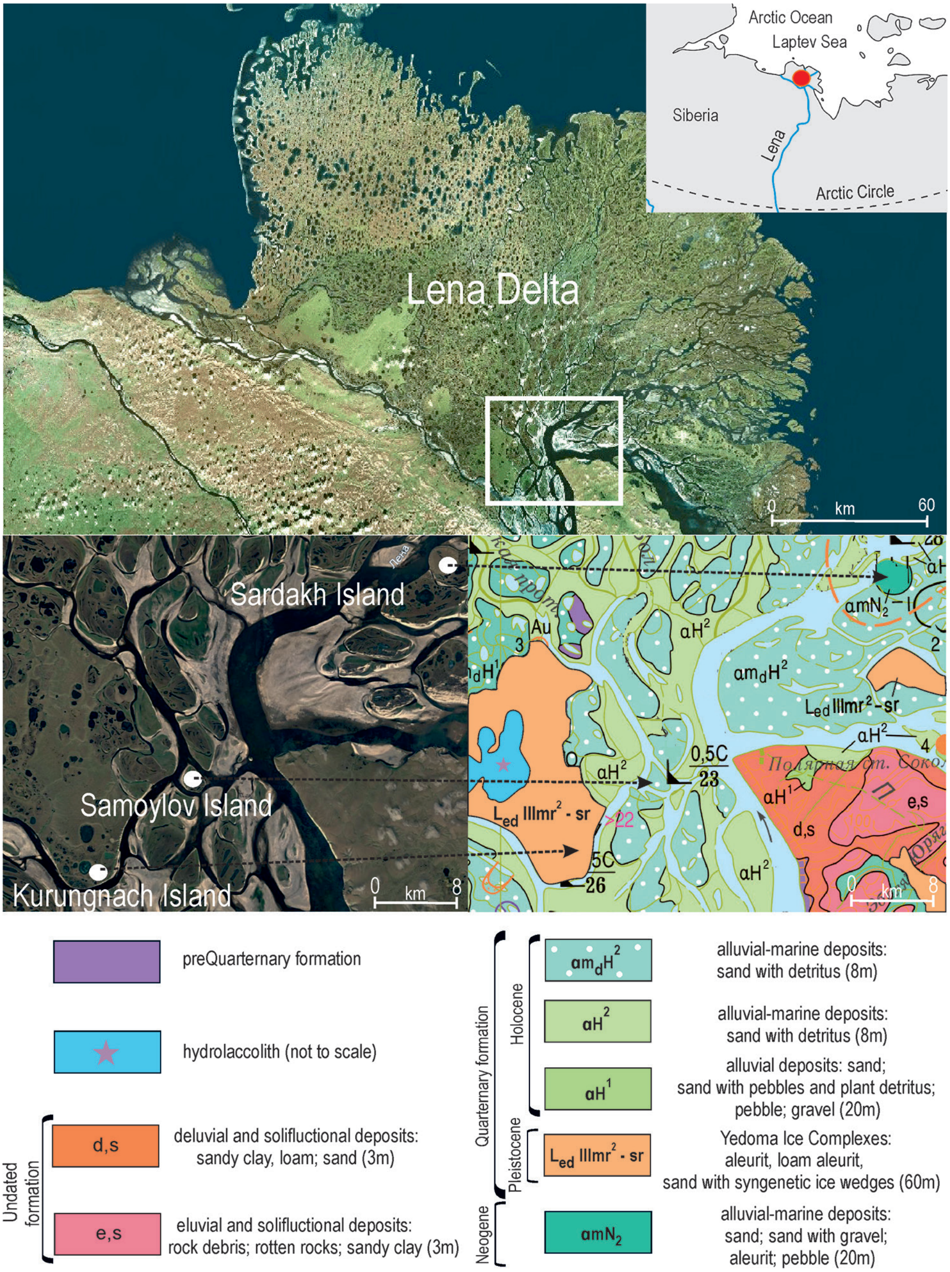

Fig. 1. The area studied [16]
Рис. 1. Территория исследования [16]

Furthemore, the ongoing thermokarst processes can also influence them by releasing chemical elements from the permafrost deposits present under the lake bottom [3, 4]. The lake sediments constitute the terminal accumulation matrix of chemical substances in the aquatic ecosystems. However, the accumulated chemical elements can be released from the sediment to the water column if there are changes in the physicochemical parameters of the environment. In this context, two aspects of trace element content in the sediments are important to analyze: the total concentration and chemical fractions of the elements [5–7]. The assessment of the total content of the elements is necessary for understanding their regional background concentrations in the sediments [8]. However, the migration mobility of metals depends on the type of chemical bonding of the elements with the mineral and organic components of the sediments [9, 10]. Humic substances, clay minerals, and Fe/Mn hydroxides are the main accumulation matrices of elements in lake sediments. They form the most stable compounds with metals. However, metal ions can be associated with sediments by weak chemical bonds. The most mobile fractions are predominantly involved in the biogeochemical cycles. Therefore, comprehensive geochemical surveys are necessary to understand the specific features of the processes of microelement migration in the permafrost-affected limnic systems.

This research is a continuation of the previous geochemical investigation of the permafrost-affected lakes of the Lena Delta in the vicinity of the Samoylov station [11]. The aim of this paper is to estimate the role of organic and mineral components of sediments in the accumulation of the trace elements (V, Cr, Co, Ni, Cu, Zn, Sn, and Pb). Importantly, it is the first full-spectrum analysis of the chemical fractions of metals in lake sediments of the region studied that has ever been conducted.

MATERIALS AND METHODS

Study area

The delta of the Lena river is the largest in the Arctic. Its Quaternary sediments overlay the Ordovician–lower-Carboniferous terrigenous-carbonate deposits (Fig. 1). Three main geomorphological terraces have been distinguished in the Lena Delta, above the floodplain [12, 13]. The lakes studied are located on the first terrace (Samoylov Island), the third terrace (Kurungnakh Island), and on the erosional residual outcrop (Sardakh Island). The first terrace is Holocene in age (3–4 ka BP) and is represented by sandy deposits with plant remains (moss detritus). The third terrace is composed of late Pleistocene (13–12 ka BP) ice-bearing permafrost deposits — Yedoma Ice Complexes [14]. The Sardakh Island is made up of Miocene and Pliocene deposits. Furthermore, on the island, the crystalline basement lies at a depth of about 100 m under Neogene deposits [12]. The lakes of the Lena Delta developed predominantly under thermokarst conditions [15]. The main types of water bodies are polygonal ponds, deep thermokarst lakes and ox-bow lakes [4].

Sampling procedure

Samples were taken from 10 small lakes (their areas do not exceed 0.05 km^2) in August 2019 (Table 1). The hydrochemical features of the lakes of the Lena Delta can be found in a work by Chetverova et al. [3]. The surface sediment (0–10 cm) was collected using the Van Veen grab. Samples were taken at five locations within each lake, chosen in such a way as to cover the whole variety of depths of the water body. They were subsequently combined to form an integral sample for each given lake.

Chemical analysis of the sediments

For the analysis of acid-soluble forms of the trace metals, fully dried sediment subsamples were decomposed with a mixture of HNO_3 (70 %, high purity GOST 11.125–84), HCl (38 %, high purity, GOST 14.261–77) and H_2O_2 (33 %, analytical grade, GOST 10929-76). This method of acid decomposition allows extracting most of the

Table 1

The characteristics of the lakes of the Lena Delta that were studied

Таблица 1

Характеристики изученных озер дельты р. Лены

Characteristics	Lake 1	Lake 2	Molo Lake	Fish Lake	North Lake	Shallow Lake	Banya 2 Lake	Banya 3 Lake	Banya Lake	Flood Lake
Latitude (°) Longitude (°)	72.57111 127.23638	72.30856 126.24546	72.37814 126.49703	72.37363 126.48692	72.38452 126.48807	72.37506 126.51104	72.37000 126.50472	72.37040 126.51721	72.36841 126.48591	72.38704 126.48091
Island	Sardakh	Kurungnakh	Samoylov							
Geomorphological terrace[1]	Residual outcrop	3rd	1st							
Type of lake	Thermokarst					Polygonal pond	Oxbow			Floodplain
Flooding regim[2]	Isolated from river influence						Affected by high seasonal river floods	Regularly affected by seasonal river floods		
The depths of sampling, m	0.5–1.5	0.5–1.1	2.6–6.2	0.5–12.7	2.5–4.8	0.5–3.1	1.0–16.3	1.0–6.3	1.0–5.2	0.3–0.6
Field description of the sediments	Grey-brown aleurite	Grey sand with aleurite	Grey argillaceous silt			Grey argillaceous silt with aleurite	Grey argillaceous aleurite	Grey argillaceous silt		
DOM (%)[3]	no data	13.5	6.3	4.0	no data	4.5	6.0	15.0	7.8	no data

Note. [1] — According to [12, 13]; [2] — According to [3]; [3] — % of dispersed organic matter in the sediments according to [9] metal ions. The exceptions are metals associated with insoluble residue (aluminosilicate compounds). However, they do not participate in biogeochemical processes, except for the mechanical movement of particles of primary minerals.

There are some fractional schemes of trace elements for soils and sediments [5, 7, 9, 17]. In this work, the following metal fractions were extracted: metal exchange forms (fraction 1), metals specifically sorbed by various components of the sediment (fraction 2), metals associated with the most stable organometallic complexes (fraction 3), metals associated with the hydrated Fe and Mn oxides (fraction 4), and the most residual (stable) phase (fraction 5), calculated by subtracting the total for the previous four fractions from the content of acid-soluble forms of the chemical element. The chemical reagents that were used for the fractional analysis are detailed in Guzeva et al. [9].

The content of trace elements in all the extractions was measured by the ICP-MS instrument (ELAN 9000, ICP-MS instrument by PerkinElmer, USA). All the analyses were performed twice. The result was accepted as correct if the difference between the values did not exceed 20 %. The parameters of the measurements are presented in Table 2. Certified reference materials (sediment sample from Lake Baikal BIL-1 — GSO 7126-94) were used for quality control.

Table 2

Parameters of ICP-MS measurement of the trace element content in the sediment samples

Таблица 2

Параметры измерения концентраций металлов в пробах донных отложений методом ИСП-МС

Element	[1]LOD	$^2\Delta_{rel}$	$^3\Delta_{abs}$
V	3.10	5.60	1.57
Cr	2.50	2.30	2.37
Co	0.03	1.10	0.54
Ni	6.20	5.50	1.20
Cu	0.68	1.90	0.67
Zn	0.51	12.00	3.99
Sn	0.10	19.10	0.02
Pb	0.21	4.40	0.47

Note. [1]LOD — limit of detection (mg/kg); $^2\Delta_{rel}$ (%) — the relative measurement error; $^3\Delta_{abs}$ — the absolute measurement error (mg/kg).

Data processing

The content of acid-soluble forms of the metals was statistically summarized using the maximum, minimum, and average values as well as the coefficient of variation (CV, %). It is also important to note that, even though the said content is close to the total amount, it can be lower. There was not enough data about the local geochemical background for the lake sediments of the Lena Delta, therefore clarke values for the continental crust [18] were used as the reference.

The assessment of the degree of risk to the aquatic ecosystem, in terms of the mobility (bioavailability) of metals, was carried out using the indicator of the risk of secondary water pollution (RAC index) [19]. This index takes into account the strength of the bond of metals with the sediment components (the sum of 1 and 2 fractions). According to this indicator, there are five degrees of risk: < 1 % — no risk, 1–10 % — low degree, 10–30 % — medium, 30–50 % — high, and > 50 % — very high.

RESULTS

Trace metal content in the sediments

The details of the content of acid-soluble forms of the trace metals and its comparison with the clark values are presented in Table 3. Cr, Co, Ni, Cu, Zn, and Pb are distributed quite homogeneously in sediments of different lakes from the area studied. For most of them, the value of the coefficient of variation does not exceed 33 %. The exceptions were V and Sn. Their concentrations in the sediments of polygonal Lake Shallow exceed the average values twice and thrice, respectively. Nevertheless, the concentration of all the elements is either lower or at the same level compared to the clark values.

Table 3

The content (mg/kg) of acidic-soluble forms of the trace elements in the lake sediments of the Lena Delta

Таблица 3

Содержание кислоторастворимых форм металлов (мг/кг) в пробах донных отложений озер дельты р. Лены

Lake/Element	V	Cr	Co	Ni	Cu	Zn	Sn	Pb
Lake 1	27.37	41.55	13.61	26.79	16.22	81.11	0.09	11.16
Lake 2	31.45	40.85	8.87	17.90	10.49	56.81	0.14	7.07
Molo Lake	29.80	43.32	12.98	23.10	13.03	81.75	0.87	9.70
Fish Lake	38.14	51.35	12.39	27.04	16.86	94.48	0.54	11.17
North Lake	23.27	39.98	9.79	20.85	10.89	76.40	0.20	8.07
Shalow Lake	78.20	45.07	9.28	22.90	12.20	72.10	1.43	9.22
Banya 2 Lake	34.93	48.18	11.01	23.56	14.78	83.48	0.42	10.16
Banya 3 Lake	32.68	46.90	10.29	22.27	13.88	81.42	0.28	9.08
Banya Lake	30.44	46.82	10.78	23.27	13.57	84.41	0.40	9.15
Flood Lake	31.09	43.71	11.45	23.37	15.10	84.60	0.27	10.94
Max	78.20	51.35	13.61	26.79	16.86	94.48	1.43	11.17
Min	23.27	39.98	8.87	17.90	10.49	56.81	0.09	7.07
Average	35.74	44.77	11.05	23.11	13.70	79.66	0.46	9.57
Clark[1]	106.00	92.00	15.00	50.00	27.00	75.00	2.50	17.00
CV (%)[2]	43	8	14	11	15	12	88	14

Note. [1]According to [20]; [2] CV — coefficient of variation.

Fractionation of trace metals in the sediments

The percentage of different fractions of the metals, broken down into individual lakes, is presented in a set of bar plots (Fig. 2). Most of the metals are predominantly associated with residual compounds (fraction 5) in all the lake sediments studied. All the investigated elements were identified in organometallic complexes (fraction 3) and forms associated with Fe/Mn hydroxides (fraction 4). Fraction 4 is most significant for Cr and Zn in all the lakes, and also for Co in Lake 2, North Lake, Banya Lake, Molo Lake as well as Shallow Lake. For V, Ni, Sn, Pb, and Cu, this phase does not exceed 1–3 %.

Furthermore, all the metals analyzed were observed in specifically sorbed forms (fraction 2). Co, Cu, Cr, and Ni are mostly associated with this phase. In exchange forms (fraction 1) all the metals were also identified in all sediment samples. The exception was Zn. Its concentration in this phase was lower than LOD (Shallow Lake, Molo Lake, Flood Lake, and Fish Lake), did not exceed 1–3 % (Lake 1, North Lake, Banya Lake, Banya 2 Lake, and Banya 3 Lake) or was 10 % (Lake 2).

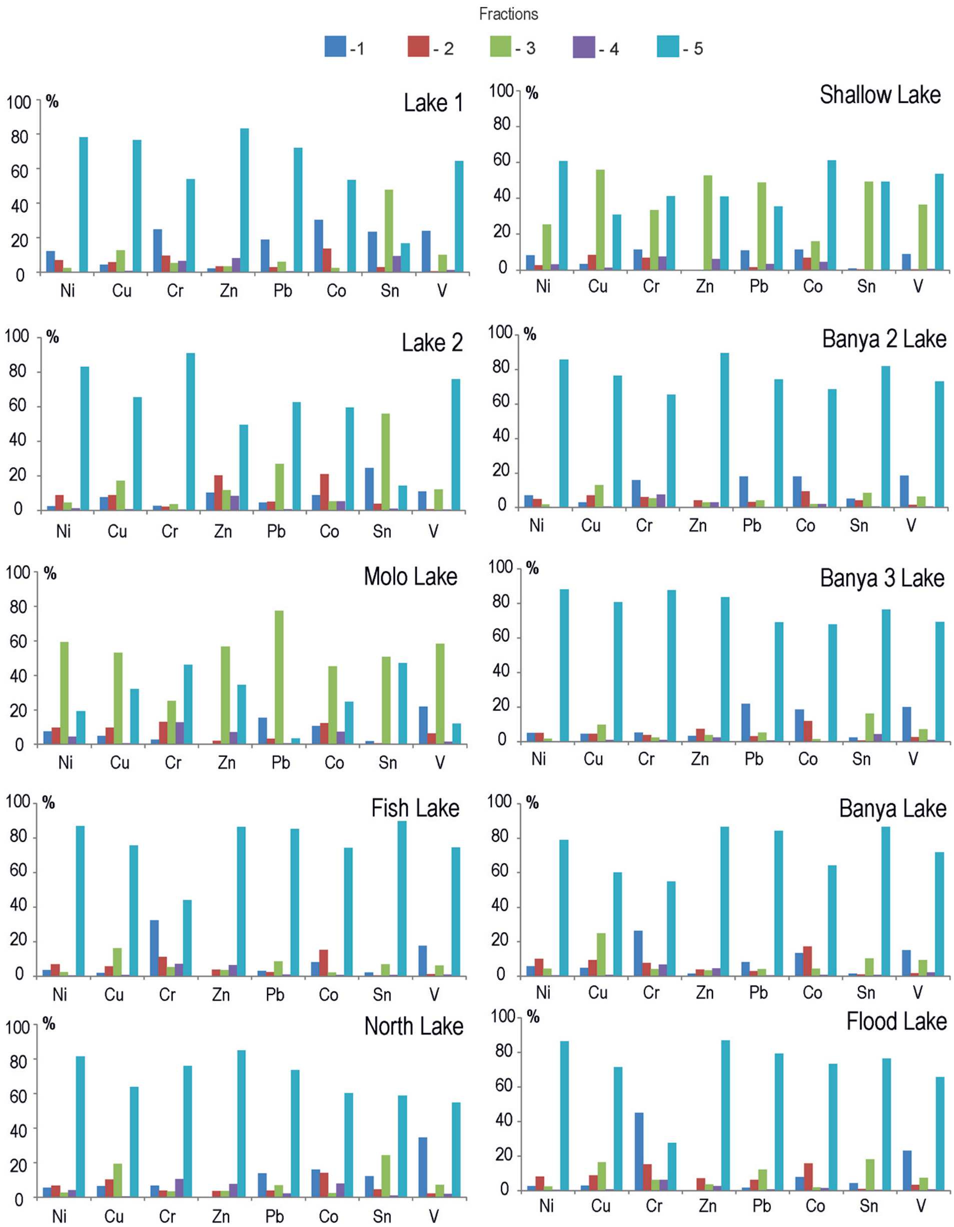

Fig. 2. The fractions of the metals (% of acid-soluble forms) in lake sediments of the Lena Delta: fraction 1 — exchangeable ions; fraction 2 — specifically sorbed; fraction 3 — organometallic complexes; fraction 4 — metals, associated with Fe/Mn hydroxides; fraction 5 — residual compounds

Рис. 2. Фракции металлов (% от кислоторастворимых форм) в озерных отложениях дельты Лены: фракция 1 — обменные ионы; фракция 2 — специфически сорбированная; фракция 3 — металлоорганические комплексы; фракция 4 — металлы, связанные с гидроксидами Fe/ Mn; фракция 5 — остаточные соединения

The assessment of the mobility of the elements is presented in a separate bar chart (Fig. 3). According to the RAC index, the risk of secondary pollution for Ni, Cu, Zn, Sn, and Pb does not exceed the medium level. However, for V a high level is shown in the sediments of North Lake; for Cr — in Banya 2 Lake, Lake 1, and Fish Lake; for Co — in Lake 1. Cr has a very high level of RAC (60 %) in the sediment of Flood Lake.

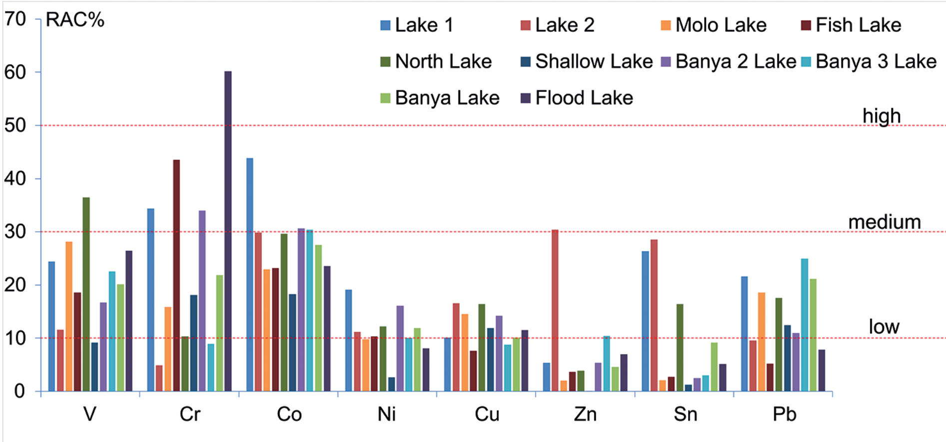

Fig. 3. RAC, (%) index — risk of secondary water pollution (sum of fractions 1 and 2). Red dotted lines indicate the level of the risk

Рис. 3. RAC (%) индекс — риск вторичного загрязнения (сумма фракций 1 и 2). Красная пунктирная линия демонстрирует уровень риска

DISCUSSION

The previous hydrochemical study in the Lena Delta revealed differences in the content of the main ions and some trace elements in the water of lakes that are characterized by different origin, flooding regimes and geological basement of the catchment area [3]. However, our work did not reveal any similar difference in the content of the majority of metals analyzed in the lake sediments. Futhermore, the elements are predominantly associated with residual compounds (crystal structure of the minerals). Therefore, the content of trace elements in all the lakes studied is determined by natural (lithogenic) sources [21]. The metals are distributed quite homogeneously in the sediments of different lakes from the area studied and the concentration of all the elements is either lower or at the same level compared to the clark values. Thus, Quaternary (Samoylov and Kurungnach Islands) and Neogene (Sardakh Island) deposits that constitute the catchment areas of the lakes studied have a similar geochemical composition. The trace elements entering with river water do not influence the total content of the trace elements in the sediments of different lakes. The pristine character of the Lena River water in terms of trace metal concentrations was revealed previously [22], but more recent data are limited. Also, it should be noted that the impact of Samoylov station on the accumulation of the trace elements in the lake sediments of the island was not observed.

The Lena Delta lakes studied were characterized by argillaceous sediments depleted in dispersed organic matter [11]. Despite this fact, stable organometallic compounds of the metals were identified in all the lakes studied (fraction 3). Humic acids of this fraction form stable chelate complexes with metals, reducing the potential bioavailability [23]. In this work, the phase is predominant in the sediments of Lake Molo and Shallow for most of the metals analyzed both in relative and absolute content. However, previous research of the sediment humic acids of the lakes studied showed their generally low maturity degree and low potential complex-forming ability [11]. But among the water bodies studied, the molecules of the acids in Molo Lake and Shallow Lake are the most hydrophobic and resistant to oxidation. Thus, they can form relatively more stable compounds with metals. Generally, the formation of complex compounds of metal ions with organic substances (humic and fulvic acids) is one of the dominant mechanisms in their fixation in aquatic and terrestrial environments [11, 24, 25]. In our work, humic matter binds predominantly Cu, Sn, and Pb. Similar geochemical behavior of these elements was observed in the non-permafrost lakes of the other regions of the Arctic zone [11, 26]. However, the sediments in these lakes contain more organic matter which can bind up to 70–80 % of the total content of some metals.

Metals associated with hydrated oxides of Fe and Mn also belong to the most stable compounds. Fe/Mn oxides are significant absorbers of the metals [9, 25, 27] and can be present in sediments as nodules, cement, or films on solid particles. Metal ions of this fraction can enter the lake water when sediment conditions change into the reducing environment (Eh decrease). However, the role of the oxidized form of Fe and Mn in the sorption of trace metals in the lake sediments studied is low. It could be explained by the reducing conditions of the sediments. The presence of hydrotroilite (black inclusions) was noted in the sediments during the sampling (Table 1). This mineral ($Fe(HS)(OH)\cdot x\ H_2O$) indicates the anoxic environment [28].

Fractions 1 and 2 are the most unstable. They are used to assess the potential mobility (bioavailability) of elements in aquatic ecosystems. Specific sorbed ions (fraction 2) are bonded with sediment components by non-ionic bonds (non-exchange form). Metals of this phase are part of the surface complexes and precipitates. The fraction also includes carbonate forms of metals and metals associated with labile organic matter. This type of compounds is in an intermediate position between the most mobile fractions (exchangeable) and relatively stable ones. The significance of this fraction for a number of elements is probably explained by the predominance of carbonate and terrigenous-carbonate rocks in geological formations of the Lena Delta and high hydrophilicity (lability) of humic molecules [11]. The metal ions of this fraction can desorb with pH decrease. The exchangeable forms (fraction 1) include the most mobile ions. The fraction is sensitive to changes of water ionic composition in terms of release of the metals in the water.

In general, some elements are characterized by high mobility because they are released as part of readily soluble compounds from the thawing permafrost of the catchment area or Lena river water during flooding. Furthermore, the high-oxygen (hydrophilic) humic acids of the sediments could form water-soluble complexes with metals [11]. This contributes to mobilization of the elements from the sediments.

CONCLUSION

The results of the study showed the relatively homogeneous distribution of the trace metals (V, Cr, Co, Ni, Cu, Zn, Sn, and Pb) in the sediments of different lakes of the Lena Delta. Most of the elements are predominantly fixed in the lattice structure of residual minerals and stable organometallic compounds.

According to our fractional analysis of the metals and the results of previous research of humic acids, the organic matter and mineral components (hydrated Fe/Mn oxides) of the sediments studied have a relatively low potential ability to fix metals into stable complexes. Thus, if the trace elements enter into aquatic ecosystems as part of readily soluble compounds, they can become bioavailable due to physical and chemical changes in the aquatic environment. Thus, there is a risk of secondary pollution of water from the sediments, and the most mobile elements are V, Cr, and Co.

Therefore, the geochemical results of this work reveal high sensitivity of the limnic system of the region to potential anthropogenic impact and climate change (releasing of chemical elements due to permafrost degradation). So, the data obtained can serve as reference values of trace metal concentration in sediments for further study of the permafrost-affected lakes of the region. Furthermore, the findings are important for developing a relevant monitoring system for the lakes of the Arctic zone.

Competing interests. No conflicts of interest involved.

Конфликт интересов. Конфликт интересов отсутствует.

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Фракции металлов в отложениях озер зоны многолетней мерзлоты севера Сибири, дельта р. Лены

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Резюме

Цель исследования — оценка роли компонентов донных отложений в накоплении микроэлементов в озерах криолитозоны (дельта Лены, северная Сибирь). Проведен фракционный анализ некоторых микроэлементов (V, Cr, Co, Ni, Cu, Zn, Sn, Pb). Пробы были отобраны из 10 озер в летний период экспедиции «ЛЕНА 2019». Содержание химических элементов в осадках измеряли прибором ICP-MS. Результаты работы показали относительно однородное распределение кислоторастворимых форм металлов в донных отложениях разных озер. Большинство элементов преимущественно закреплено в устойчивых минеральных и металлоорганических соединениях. Однако для V, Co и Cr в некоторых озерах отмечен высокий уровень подвижности. Металлы растворимых комплексов способны мигрировать из донных отложений в воду за счет изменений физико-химических параметров водной среды. Кроме того, органические и минеральные вещества осадков обладают низкой потенциальной способностью связывать металлы в устойчивые соединения.

Ключевые слова: дельта Лены, многолетняя мерзлота, озерные отложения, Сибирь, фракции металлов.

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Фракции металлов в отложениях озер зоны многолетней мерзлоты севера Сибири, дельта р. Лены (расширенный реферат)

Дельта р. Лены располагается на севере Сибири в области развития многолетней мерзлоты. На данной территории сосредоточено большое количество озер, на гидрологические и гидрохимические особенности которых преимущественно влияют поверхностные стоки с водосборных площадей, подземные воды сезонно-талого слоя, а также паводковый режим реки Лены. Кроме того, химические элементы могут мигрировать в воды озер из многолетнемерзлых отложений, подстилающих котловину озера, из-за протекающих на территории термокарстовых процессов. Так как донные отложения представляют собой терминальную матрицу накопления химических веществ в аквальных экосистемах, их комплексные геохимические исследования необходимы для понимания процессов аккумуляции и миграции микроэлементов в криолитозоне.

Цель работы — оценка роли органических и минеральных компонентов озерных отложений в накоплении микроэлементов (V, Cr, Co, Ni, Cu, Zn, Sn, Pb). В задачи исследования входил анализ всего спектра фракций металлов в пробах донных осадков.

Донные отложения были отобраны из 10 малых озер трех островов дельты р. Лены, которые характеризуются различным геологическим строением и режимом затопления речными водами. Кроме того, при выборе объектов исследования были приняты во внимание основные типы озер территории: небольшие полигональные, глубокие термокарстовые и старичные озера. Были исследованы следующие формы нахождения металлов: обменные ионы (фракция 1); специфически сорбированные металлы (фракция 2); металлоорганические комплексы (фракция 3); металлы, связанные с гидратированными оксидами Fe и Mn (фракция 4); остаточная (устойчивая) фаза (фракция 5), рассчитанная путем вычитания суммы предыдущих четырех фракций из содержания кислоторастворимых форм элемента.

В результате сравнения концентраций кислоторастворимых форм металлов в отложениях разных озер было показано, что четвертичные и неогеновые отложения, слагающие водосборы водных объектов, характеризуются сходным геохимическим составом. Кроме того, процесс затопления озер речными водами не влияет на валовое содержание металлов в донных осадках озер. Следует также отметить, что, по полученным данным, воздействия функционирования научной станции о. Самойловский на накопление микроэлементов в озерных отложениях острова не наблюдается.

Результаты фракционного анализа свидетельствуют о том, что металлы в донных отложениях преимущественно связаны с кристаллической структурой минералов, попадающих в озера с водосборных бассейнов. Следовательно, содержание микроэлементов в озерах определяется природными (литогенными) источниками.

Глинистые отложения изученных озер характеризуются невысокими содержаниями рассеянного органического вещества, тем не менее металлы в органической фракции были идентифицированы во всех пробах. Устойчивые (хелатные) органоминеральные соединения являются особенно важной формой металлов в отложениях двух озер, характеризующихся наиболее зрелыми (конденсированными) молекулами гумусовых кислот. В осадках других озер преобладают алифатические гидрофильные гуминовые кислоты, которые способны образовывать легкорастворимые комплексы и соли с металлами, мобилизуя элементы из отложений. Роль гидратированных оксидов Fe и Mn в сорбции микроэлементов в отложениях исследованных озер невелика, что можно объяснить восстановительными (бескислородными) условиями среды.

В некоторых озерах была выявлена высокая потенциальная геохимическая подвижность (биодоступность) V, Cr и Co. Обменные и специфически сорбированные формы металлов переходят в водную толщу даже при незначительных изменениях физико-химических параметров среды. Наличие данных неустойчивых форм может быть связано, с одной стороны, с изначальным попаданием металлов в экосистему озера в составе легкорастворимых соединений из оттаивающих многолетнемерзлых пород водосбора или с речными водами р. Лены при половодье. С другой стороны, гумусовое вещество и гидратированные оксиды Fe/Mn исследованных отложений, как показывают результаты, обладают низкой потенциальной способностью связывать металлы в нерастворимые комплексы.

Полученные данные могут служить референтными значениями концентрации V, Cr, Co, Ni, Cu, Zn, Sn, Pb в донных отложениях озер и необходимы для оценки роли гумусового вещества в накоплении и миграции металлов в озерах региона в условиях меняющегося климата (деградация многолетней мерзлоты). Кроме того, результаты работы важны для разработки научно обоснованной системы мониторинга озер Арктической зоны РФ.