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Uranium, Lithium, and Arsenic in Salt Lakes of Eastern Transbaikalia

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Abstract

Distribution analysis of lithium, uranium, and arsenic in water of mineral lakes of Eastern Transbaikalia that are different by mineralization degree, macrocomponent composition, and water pH was performed. Lithium is mainly concentrated in more mineralized lakes, and arsenic and uranium – in less saline, *i.e.* soda, as established. According to thermodynamic calculations, lithium is mainly present as Li^+ ion; LiCO_3^- acts as second biggest in soda water bodies, and LiCl – in chloride. Uranium is mainly found as uranyl tricarbonate ($[\text{UO}_2(\text{CO}_3)_3]^{4-}$) and dicarbonate ($[\text{UO}_2(\text{CO}_3)_2]^{2-}$). Arsenic in oxidative environment of lakes is presented by hydroarsenate (HAsO_4^{2-}), and in reductive hydrosulphuric – by dihydrogenarsenite (H_2AsO_3^-). Lithium does not form its own minerals in lakes. Reducing environment, in which uranium is precipitated as uranium hydroxide ($\text{U}(\text{OH})_4$), may be a geochemical barrier for uranium. The formation of sodium uranospinite ($\text{NaUO}_2\text{AsO}_4$) that may limit arsenic content in water too is probable under oxidative conditions. A similar trend in the distribution of their contents in chloride lakes is observed for lithium and chlorine. Direct dependence of the concentrations of uranium and arsenic on the contents of hydrocarbonate and carbonate ions is typical of soda lakes. The lack of appropriate specialization in the catchment areas of most lakes with a wide variation of contents of lithium, uranium and arsenic in their water, and also the presence of direct dependencies on the contents of the main macrocomponents indicate the prevailing influence of geochemical environment in their accumulation, the formation of which is mainly determined by evaporative concentration of water and hydrolysis of alumino-silicate rocks. The prevalence of this or that process ensures the formation of a certain chemical type of lakes and the appropriate set of elements.

Key words: lithium, uranium, arsenic, water-rock interaction, evaporative concentration, salt lakes in Eastern Transbaikalia

INTRODUCTION

Over recent years, salt lakes are increasingly seen as sources of mineral renewable resources, exploitation of which does not require

expensive technologies and is considered to be environmentally safe [1, 2]. Earlier, salt lakes were mainly studied as objects of probable extraction of salts, primarily, soda, mirabilite, and galite [3, 4]. In Eastern Transbaikalia,

extraction of salt was carried out commercially at Borzinskoe Lake; mirabilite was mined at Shihlin Nord Lake, soda – at Bain-Tsagan and Doroninskoe Lakes. Meanwhile, numerous mineral lakes in Eastern Transbaikalia together with lakes of regions of Russia (Altai), China, Mongolia, and other countries can be considered as promising for industrial water accumulating many chemical elements, including rare earth ones [5–7, *etc.*]. In lakes of Western Mongolia, the behaviour of lithium and uranium promising for commercial recovery is discussed in detail; the main factors that contribute to their accumulation in water have been determined [8]. There is only one publication regarding this kind of information for lakes of Eastern Transbaikalia [9]. Therefore, highlighting the key factors that ensure the accumulation of uranium and often arsenic accompanying it is the purpose of this paper.

RESEARCH OBJECTS AND METHODS

Salt lakes in Eastern Transbaikalia are situated in an area with a marked sharply continental climate. From the west and the east, the territory is limited by coordinates of 115–116° east longitude, from the north – 50° latitude, from the south – by the state border with widespread continental salinization processes on basin bottoms. Uneven rocks take part in the geological structure of basins; they are more often represented from the surface by sedimentary deposits with up to 1700 m in thickness (Citino-Ingoda, Ononsk). In cavity framing, granites and gneisses are most widespread; in the area of the Torey lakes, basalts rise at the daytime surface; on the left bank of Borzuya River, a ledge of terrigenous-carbonate rocks of the Devonian rises; to the north of Onon River, Mesozoic magmatic rocks and Paleozoic metamorphic series are common [11]. The largest lakes, such as Barun and Zun-Torey have a total area of the water table up to 850 km². Other lakes are different in small sizes. They mostly become dry to the bottom during drought years, and in wet periods are again filled with water. All lakes are drainless, have small catchment areas; water nutrition of their bulk mass is atmospheric, directly at the water

area and underground during heavy rains and frequent rains – *via* surface runoff.

Sampling in lakes was carried out in 2013–2015 during expedition studies in the basins of Shilka, Argun, and Uldza rivers (in the territory of RF) (Fig. 1). In total, 90 samples of lake waters were selected and analyzed. Water at the site was filtered *via* a membrane filter with a pore diameter of 0.45 μm and acidified with exceptionally pure concentrated nitric acid of the qualification (1 : 1) to pH < 2 immediately after filtration. Samples for submission to the device were diluted with deionized water with 18.2 mΩ/cm resistance to the total content of salts of less than 0.01 g/L. Determinations of microcomponents in lake water were carried out at analytical centres of Vinogradov Institute of Geochemistry, SB RAS (Irkutsk) and Institute of Solid State Chemistry and Mechanochemistry, SB RAS (Novosibirsk) by inductively coupled plasma mass spectrometry (ICP-MS). In the first case, ELEMENT 2 device (Finnigan MAT) was utilised according to the method HCAM N 480X using certified standard solutions, such as ICP Multi Element Standard Solution XII Sol, Sol X CertiPUR (MERCK) and Combined Quality Control standard IQC-026 (NIST, USA) as standards. In the second case, Inductively Coupled Mass Spectrometer (Agilent 7500a) was used. The mass spectrometer was tuned according to Tuning Solution Agilent Part Number 5184-3566. The content of trace elements was determined by multielement standard solutions (Multi-Element Calibration Standard-2A Agilent Part Number 8500-8940).

Chemical analysis of macrocomponent composition of water has been carried out by standard methods at the Institute of Natural Resources, Ecology and Cryology SB RAS (IPREK SB RAS). The concentrations of Ca and Mg were determined by atomic absorption method in the ferrous-acetylene flame using SOLAAR 6M spectrophotometer. The flame emission method was used to determine Na and K. Potentiometry using ion-selective electrodes was used to determine O₂, F, pH, E_h, and Cl, while titration for identifying the content of CO₃²⁻ and HCO₃⁻. Sulphate ions were analyzed by the turbidimetric method as barium sulphate.

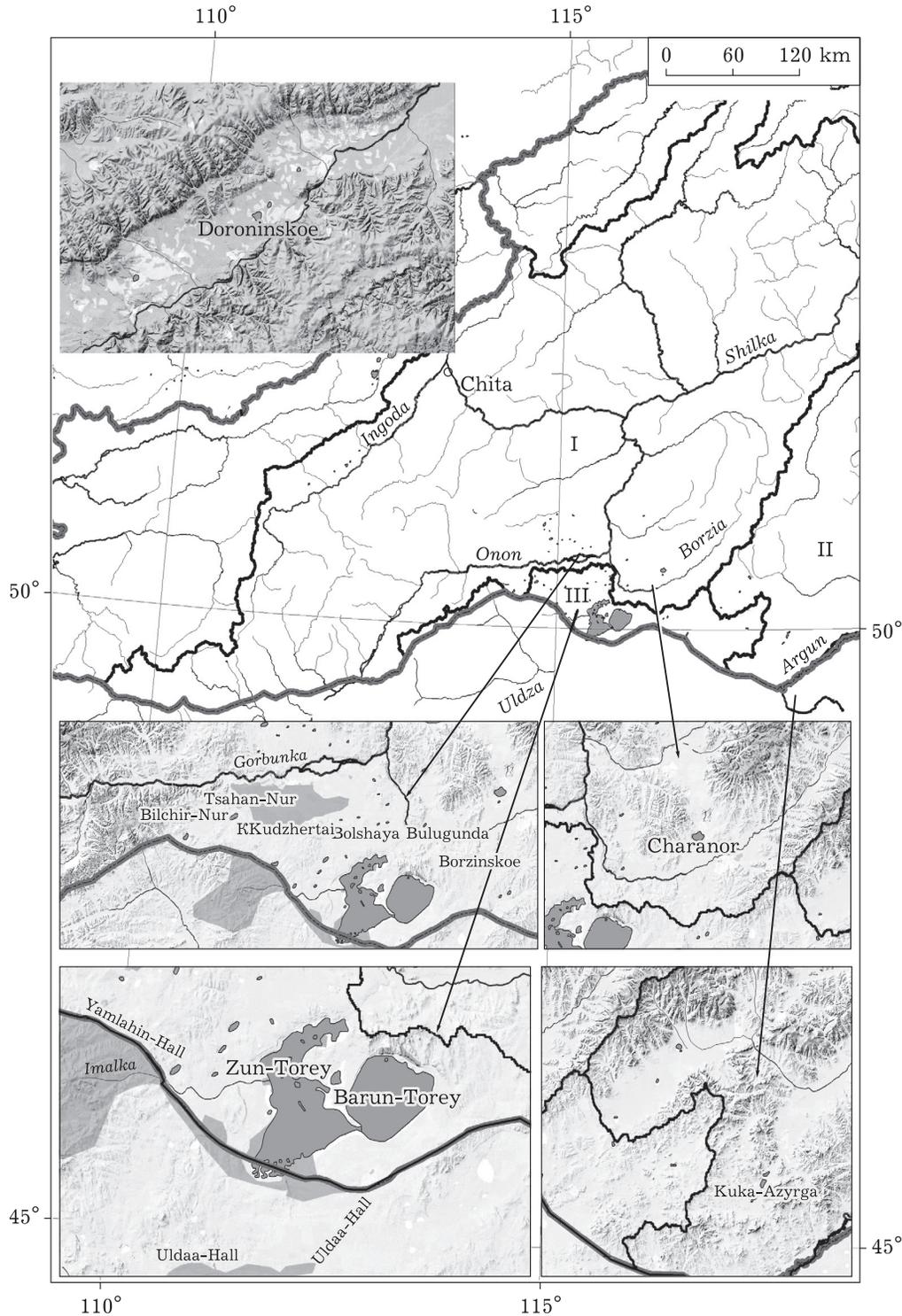


Fig. 1. Layout of mineral lakes of Eastern Transbaikalia by the highlighted river basins (I – Shilka R, II – Argun, III – the Uldza) according to Egidarev [10].

RESULTS AND DISCUSSION

Mineral lakes are mainly presented by basic type; chloride type is met much more rarely, the prevalence of sulphates has been determined in

single cases (Table 1). The brine salinity of lakes reaches 343 g/L (Borzinskoe Lake). Carbonates in lake water mainly prevail at the salinity up to 15 g/L, chlorine becomes the main anion in more salt lakes. Sodium prevails in the cation

composition, becoming the sole significant cation to determine the chemical type at a salinity of 5 g/L. The pH value of water reaches 10.7 (Kudzhertai Lake) at the average value for soda lakes of 9.54, for chloride ones – 8.4. The redox potential (E_h) of water of soda lakes varied from –380 to 380.7 mV, chloride ones – from –36.3 to 97.7 mV; negative values of E_h are driven by the development of mainly sulphate reduction.

High values of pH determine the behaviour of many elements including calcium and magnesium, the content of which on average is by an order of magnitude lower in soda

lakes. Compared to chloride ones, soda lakes are characterised by higher concentrations of U, V, Mo, Zr, and As, while Li is contained in smaller amounts. The maximum contents of As of 6 mg/L, U of 2.8 mg/L are noted in the most soda lake – Borzinskoe, however, the concentration of Li (173 $\mu\text{g/L}$) only slightly exceeds the average for the whole sample (123 $\mu\text{g/L}$). Gorbunka Lake with the maximum water mineralization of 184.4 g/L and relatively high contents of Cl, Br, Sr, Li is distinguished among chloride lakes, while the amount of U does not exceed 59 $\mu\text{g/L}$.

TABLE 1

Physicochemical characteristics of the mineral lakes of Eastern Transbaikalia

Parameters	Hydrogeochemical type of lakes					
	Soda			Chloride		
	Content			Content		
	Average	Minimum	Maximum	Average	Minimum	Maximum
	mg/L					
CO ₂	<0.44	<0.44	<0.44	144.0	<0.44	156.0
CO ₃ ²⁻	1186	16.0	18600	232.0	<0.30	840.0
HCO ₃ ⁻	1657	24.4	7930	625.9	61.2	2025
SO ₄ ²⁻	1860	10	62340	3208	210	13680
Cl ⁻	4164	28.2	133740	16531	1196	101088
F ⁻	14.0	<0.19	131.4	7.07	0.27	37.1
Ca ²⁺	13.6	0.8	80.0	93.1	6.19	617.5
Mg ²⁺	53.3	1.08	291.4	575.8	9.95	3795
Na ⁺	4853	272	124800	11662	1348	65000
K ⁺	55.8	2.3	440.8	72.3	7.5	237.6
M*	14195	1174	343227	32989	4632	184650
Si	2.2	0.3	17.7	0.7	0.4	2.0
	$\mu\text{g/L}$					
Li	87.6	2.0	3263	208.7	62.1	675.1
V	77.0	0.02	420.3	23.8	5.99	119.1
As	414.4	25.2	6007	102.9	13.6	278.9
Sr	335.9	0.04	1576	4097	129.5	18506
Br	29936	30.0	624292	78096	10580	368586
U	156.7	2.5	2818	75.6	15.6	246.1
Zr	41.3	0.12	1105	9.06	0.14	47.5
Mo	124.0	3.0	2987	88.1	15.4	378.9
E_h , mV	100	–380	381	44	–36	98
pH	9.54	9.02	10.7	8.40	7.40	9.66

Note. For the soda type of lakes, the average value is calculated according to the data of 77 samples, for the chloride type - 13 samples.

* *M is mineralization.

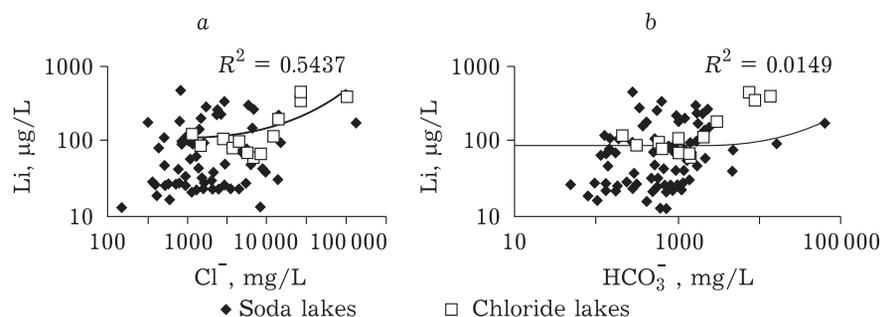


Fig. 2. Lithium content *versus* chloride (a) and bicarbonate ions (b).

To reveal causes of different behaviour of microelements, analysis in the system, such as water – rock – salts was carried out and demonstrated that the entire water was saturated with such carbonate minerals, as calcite, dolomite, halite, and gypsum. The Bil'chir-Nur Lake of chloride type, water of which is in equilibrium with gypsum is an exception. Herewith, the content of sulphates reaches 13680 mg/L , and calcium – only 160 mg/L . The water is in equilibrium with gaylussite by some soda lakes. Gaylussite formation is probable at a certain stage of soda-formation process [12]. Water equilibrium with clay minerals, mainly, montmorillonites and illites is reached, however, there is no equilibrium with primary amines, *e.g.* anorthite [13].

It is understood that calcium and magnesium are mainly bound by carbonates and aluminosilicates, potassium and sodium – by aluminosilicates, the latter – also by gaylussite. The content of other elements not bound by the secondary phase, *e.g.*, chlorine, increase with a rise of water mineralization. Therefore, chlorine accumulation process may be referred to the result of the evaporative concentration of water, and this indicates that the higher is the degree of water evaporation (with no additional chlorine source), the higher is its accumulation in lakes [14].

A single focus in content distributions of Cl and Li is exclusively registered in chloride lakes (Fig. 2, a). There is no dependence on Li with the main HCO_3^- ion in soda ponds (see Fig. 2, b). Lithium mainly migrates in water as its own Li^+ ion with the prevalence of its share in chloride lakes (Fig. 3, a). Lithium hydrocarbonate acts as second by significance in soda lakes, and LiCl – in chloride ones. Herewith, the mole fraction of associates increases with

rising concentrations of main ligands. Water saturation with lithium chloride (LiCl) and carbonate (Li_2CO_3) is not observed due to a high value of solubility products of lithium salts (see Fig. 3, b, c). Apparently, the absence of geochemical barriers allows lithium like chlorine concentrating in solutions; herewith, the higher is the concentration of chlorine, the higher are the lithium contents in lakes. The effect of evaporative processes for concentrating lithium in water is confirmed by numerous patterns: salars of Argentina, China, and Chile, Red sea, salt lakes of the Altai, *etc.* with the content up to several g/L of Li in their water [2, 15].

The presence of an additional source of lithium is not excluded in a few cases; rare metal granite pegmatites may act as which, some manifestations of which are found in the study area and high concentrations of Li in mineralized lakes are found. Thus, in Tsagan-Nur Lake with water mineralization is just 4388 mg/L , Li content is 3263 $\mu\text{g/L}$. According to calculations, the equilibrium of water with the main aluminosilicate minerals, such as eucryptite ($\text{LiAl}[\text{SiO}_4]$) and spodumene ($\text{LiAl}[\text{Si}_2\text{O}_6]$) is not reached. They may hypothetically dissolve and act as an additional source (see Fig. 3, d, e). Uranium concentrations cover the range of several mathematical orders – from 2.55 to 2819 $\mu\text{g/L}$. The magnitude of uranium concentrations itself indicates the decisive role of geochemical characteristics of the medium in uranium distribution in lake water. High contents of uranium have been detected in soda lakes with a relatively high value of pH of water (Fig. 4, a). It may be concluded from here that the available anomalies of uranium in water of mineral

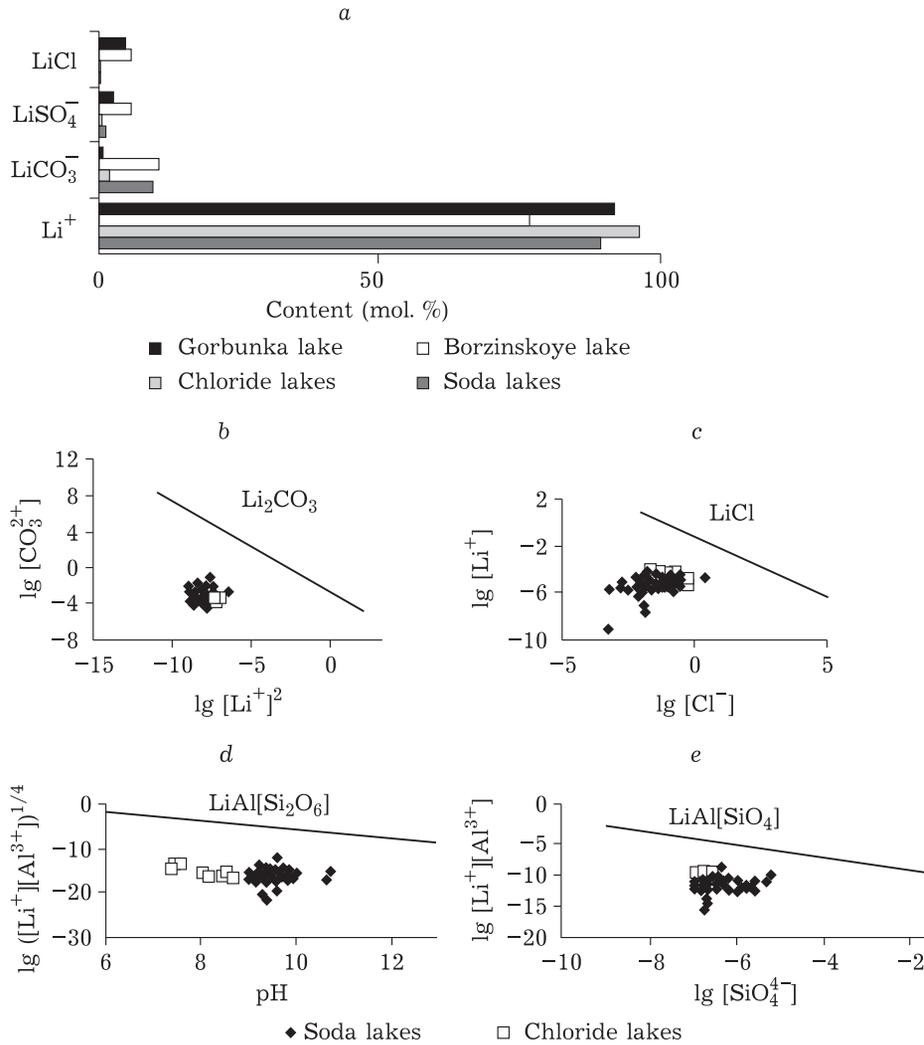


Fig. 3. Main forms of lithium migration in chloride and soda lakes (a) and water equilibrium with lithium carbonate (Li_2CO_3) (b) LiCl (c), spodumene $\text{LiAl}[\text{Si}_2\text{O}_6]$ (d), and eucriptite $\text{LiAl}[\text{SiO}_4]$ (e).

lakes of the region are not associated with ore mineralization, unlike, for example, one of the lakes of North-Western Mongolia, uranium concentration in 1.1 mg/L for which may be considered as uranium-bearing indicator [15, 16]. At the same time, rare-metal granitoids, certain intrusive complexes of which have high radioactivities, are widespread in the study area. It is natural to expect that the mineral lakes may have, in some cases, high concentrations of uranium.

According to [17] and other sources, uranium may be present as aquauranyl ($[\text{UO}_2(\text{H}_2\text{O})_6]^{2+}$), in the presence of other ligands – as complexes with them, particularly at high pH – as aquauranyls, and in case of carbonate ions – as carbonate uranyls. Out of the latter,

tricarboxylate-uranyl ($[\text{UO}_2(\text{CO}_3)_3]^{4-}$) and bicarbonate-uranyl ($[\text{UO}_2(\text{CO}_3)_2]^{2-}$) are stable in an aqueous medium (see Fig. 4, b). Tricarboxylate complex becomes prevailing at a concentration of carbonate ion of $10^{-4.5}$ M, at a concentration of carbonate of more than $10^{-3.0}$ M, the share of the complex is close to 100 %. The carbonate ion content in lakes reaches 18600 mg/L ($3.1 \cdot 10^{-1}$ mol/L), which indicates an opportunity of the formation of carbonateuranyl ions. Performed thermodynamic calculations using HydroGeo software complex confirm this [18]. Hence a direct relationship of uranium with carbonate and bicarbonate ions is clearly visible, herewith, the stability of the resulting complexes is directly dependent on the pH of the solution (see Fig. 4, c). A decrease in the pH of water

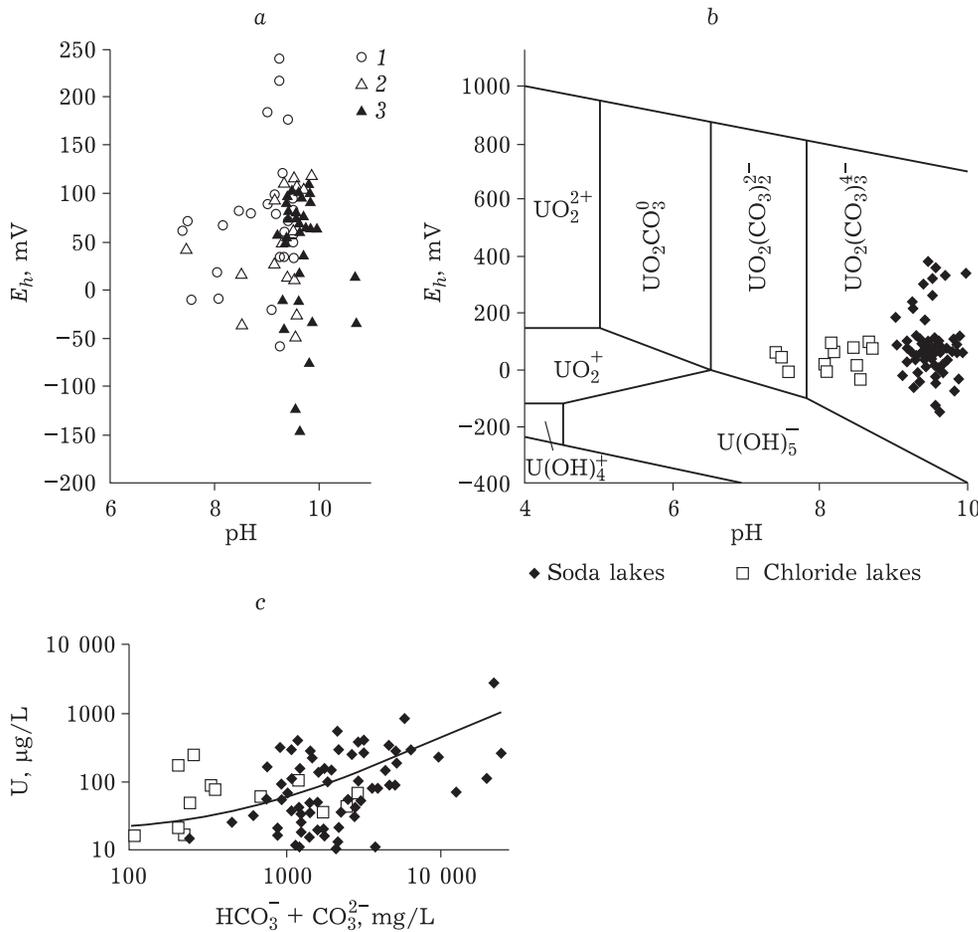


Fig. 4. Uranium content depending on pH and E_h of the medium (a), main forms of its migration (b), and also uranium contents versus carbonate complex ($HCO_3^- + CO_3^{2-}$) in water of salt lakes (c).

promotes the formation of dicarbonate-uranyl ions in chloride lakes, the presence of which directly depends on the presence of the carbonate ligand in water.

Uranium accumulation at soda-formation stage is confirmed by a bond of uranium with sodium ($r = 0.95$), since prevailing sodium concentrations compared to the rest of anions (calcium, magnesium, and potassium) in water starts from the moment of precipitation of carbonate minerals and clays [14].

A decrease in uranium concentrations at a high content of CO_3^{2-} is noted in a number of cases in soda lakes (Doroninskoe, Kudzhertai lakes, etc.), which happens in a reducing environment ($E_h = -380$ mV in water column of Doroninskoe Lake) and is probably caused by a transition of U^{6+} into U^{4+} followed by its precipitation as uranium hydroxide ($U(OH)_4$),

the solubility product (SP) of which is given [19] and equal to $1.1 \cdot 10^{-52}$, or as uranium oxide (UO_2). Precipitation of four-valent uranium oxide is probable resulting from destruction of carbonate complexes and depending on total mineralization and water composition, pH value, carbonate content begins at decreasing E_h to the range from 0 to -0.2 mV [20]. Lake water is in an equilibrium with sodium uranospinite ($NaUO_2AsO_4$), SP of which is $1.33 \cdot 10^{-22}$ (the value from the paper [19]). According to calculations, saturation with respect to autunite ($Ca(UO_2)_2(PO_4)_2$) is not reached (SP $5.5 \cdot 10^{-46}$ according to [20]). An opportunity for reduction of U^{6+} in chloride lakes in the considered cases is less probable, as oxidative conditions prevail here more often; E_h values do not decrease lower than -35 mV (Bolshaya Bulugunda Lake).

Uranium flow in soda lakes due to dissolution

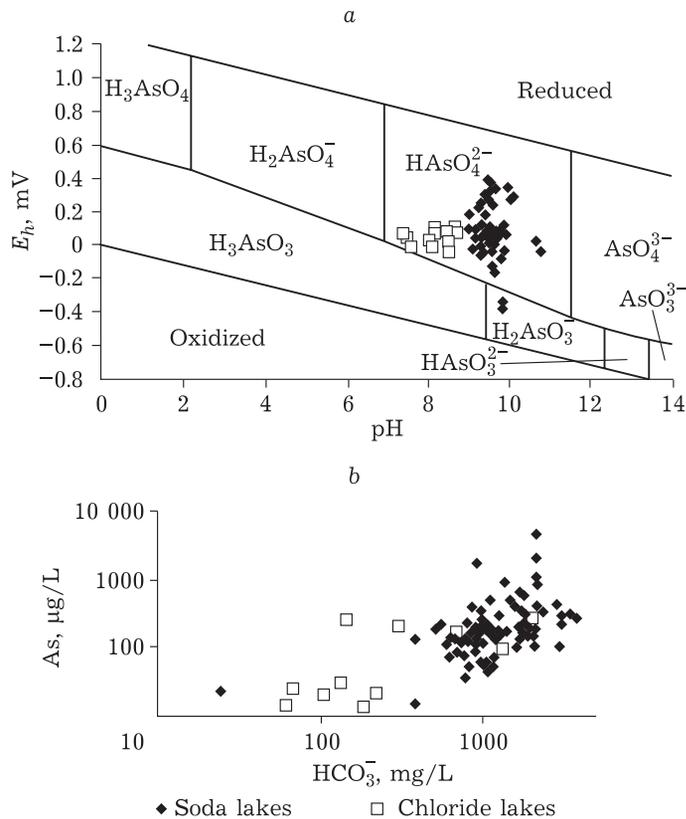


Fig. 5. Main forms of arsenic migration depending on pH and E_h conditions (a) and HCO_3^- contents in lakes (b).

by water in catchment areas and in lakes containing rocks is confirmed by the high correlation dependence of U with accompanying Zr (0.90), Mo (0.84), and V (0.87), the contents of which are much higher as by average, and maximum assessments. There are no these dependencies in chloride lakes.

According to [14], the more intensive is the hydrolysis process of aluminosilicate rocks, the higher is water alkalinity and the greater is the supply of chemical components. However, chlorine contents also increase in soda lakes with a relatively high value of pH resulting in an additional contribution of evaporation into uranium accumulation.

Arsenic behaves similarly to uranium, even though the latter is present in both types of water mainly as $HAsO_4^{2-}$ with an exception of lakes with the hydrosulphuric environment of the water column, where arsenic reduces to As^{3+} and is already present as $H_2AsO_3^-$ (Fig. 5, a). A mixture of arsenic with geochemical medium appears *via* a consistent increase in the contents of As and HCO_3^- (see Fig 5, b) and a high

correlation coefficient ($r = 0.66$). Considering that the presence of carbonic acid (carbonate and hydroxycarbonate ions in water is ensured by the $OH^- + CO_2 \rightarrow HCO_3^-$ reaction, and water itself [14] is a source of OH^- in the hydrolysis reaction, the more intense is the dissolution of minerals (subject to CO_2 supply to the system), the more is accumulated HCO_3^- . Obviously, these dependencies indicate a greater role of hydrolysis reactions in arsenic accumulation.

In some cases, one may consider desorption processes, as a result of which partial dissolution of iron oxyhydroxides on the boundary of a change from oxidizing conditions to reducing conditions leads to the liberation of chemical elements bound with them including arsenic. However, as a rule, arsenic concentrations in the hydrosulphuric environment do not exceed dozens of $\mu\text{g/L}$, therefore, this process does not yield substantial contribution to As accumulation. The same trend in uranium and arsenic distribution in soda lakes (Fig. 6, a) may also be explained by an opportunity to form joint minerals, *e.g.* above mentioned sodium

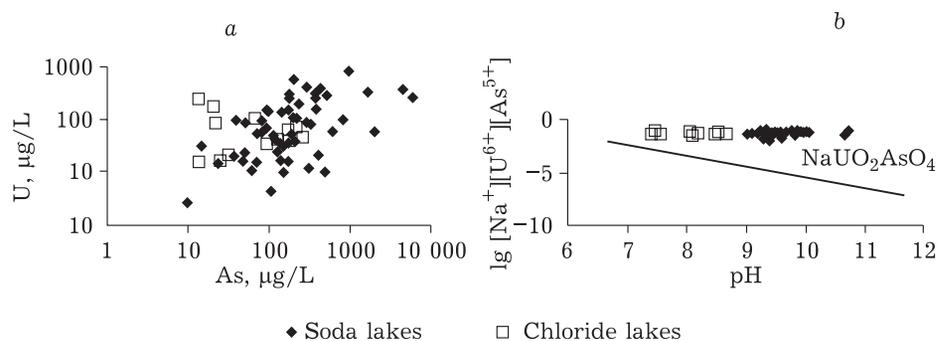


Fig. 6. Distribution of arsenic and uranium (a) and equilibrium of water with sodium uranospinite ($\text{NaUO}_2\text{AsO}_4$) (b) in soda and chloride lakes.

uranospinite, water equilibrium with which is reached in the highlighted types of lakes (see Fig. 6, b).

CONCLUSION

Distribution analysis of the concentrations of lithium, uranium, and arsenic has demonstrated that the contents of these elements in lakes of Eastern Transbaikalia reach a few milligrams and are close to industrial concentrations for their extraction. Lithium is accumulated in more mineralized chloride lakes, and uranium and arsenic – in more alkaline water. Despite the fact that Transbaikalia is the largest lithium-bearing (Zavitinskoe, Eticsinscoe, etc.) and uranium-bearing (Streltsovskoe, Berezovskoe, Orlovskoe) provinces, the considered area is located outside these deposits. Arsenic deposits (Zapokrovskoe, Klichka) are also situated at a considerable distance from the study objects. The lack of uranium, lithium, and arsenic specializations by the majority of catchment areas of lakes eliminates an opportunity to consider it the main cause of high contents of appropriate elements. Significant variations of the concentrations of these elements in the highlighted types of lakes indicate the important role of the hydrogeochemical environment in their accumulation. At the same time, the formation of the medium itself is determined by the prevalence of the evaporation process for chloride lakes, and hydrolysis of aluminosilicate minerals for soda ones, leading to the difference in accumulation of certain trace elements.

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