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SEASONAL VARIABILITY IN CHEMICAL COMPOSITION OF ANTHROPOGENIC CRYOPEGS WITHIN YAKUTSK CITY

N.A. Pavlova, M.V. Danzanova

Melnikov Permafrost Institute, SB RAS, 36, Merzlotnaya str., Yakutsk, 677010, Russia; na-pavlova@yandex.ru

Long-term and seasonal variations in major- and trace-element compositions of groundwaters in anthropogenic cryopegs stripped by boreholes in unconsolidated alluvial sediments have been monitored for thirty years at a site within Yakutsk city. Suprapermafrost cryopegs became less saline for the past five to seven years as a result of climate change in many previous years. The obtained data on chemistry and flow dynamics of groundwaters indicate enhanced hydraulic connectivity of cryopegs in the active layer. The ongoing ground temperature warming will lead to general decrease in the contents of highly soluble salts in suprapermafrost cryopegs and to increase in trace element abundances in pore waters.

Suprapermafrost cryopeg, intrapermafrost cryopeg, water chemistry, major and trace elements, long-term regime

INTRODUCTION

Urban territories in Central Yakutia are incurring environmental risks from salinization of permafrost, water-logging, and formation of cryopegs caused by anthropogenic loads. The situation is especially urgent in Yakutsk city where ever more buildings experience uneven basement settling and become dangerous [*Garagula and Ershov, 2000; Stepanov et al., 2014*]. The contents of total dissolved solids (TDS) in pore waters within the city is the highest in the oldest neighborhoods which have been exposed to the anthropogenic impact for more than 300 years [*Torgovkin and Makarov, 2015*]. TDS reach 1 % in the active layer [*Makarov, 2011*] and vary from 1 to 4 g/L in suprapermafrost groundwaters that form in the active layer in warm seasons, or even from 6 to 9 g/L in the case of impeded drainage [*Sannikova, 2005; Shepelev, 2011; Danzanova, 2016*]. The major-ion composition of these waters consists mainly of HCO_3^- and Cl^- anions, abundant Na^{2+}, less abundant Mg^{2+}, and relatively low Ca (within 20 %). Suprapermafrost waters laden in Mg and Na chlorides, which have low crystallization temperatures, remain unfrozen in winter and form cryopegs [*Anisimova, 1981; Fotiev, 2009*]. Taliks and cryopegs of different sizes and shapes are often stripped by drilling at redevelopment sites of old wooden houses in Yakutsk city, in closed basins or lakes, as well as near roadways and buried ducts [*Anisimova, 1981; Konosavskiy, 1983; Anisimova and Kurchatova, 2000*]. The water chemistry of cryopegs depends on the type and magnitude of load, as well as on the temperature and freezing conditions of the host ground. Most cryopegs have salinity about 20 g/L but may reach 35 to 200 g/L in old city parts. Such waters crystallize at –0.5 to –8.0 °C.

Long-term monitoring of negative-temperature saline groundwaters at different depths in unconsolidated alluvium within Yakutsk city has been set up in order to reveal space-time water chemistry variations of permafrost cryopegs in urban areas under the ongoing climate change.

OBJECT AND METHODS

Yakutsk city is located on a low above-floodplain terrace in the middle reaches of the Lena River. The terrace has a smooth surface, with elevations from 94 to 102 m asl [*Spektor et al., 2008*]. More than 300 m thick permafrost spreads continuously over the study area. The active layer thickness within the city is from 0.5 to 4.5 m, depending on permafrost lithology and salinity, and on surface illumination. Ground temperature and active layer thickness have shown increasing trends for the recent decades, under the effect of natural and manmade factors. The natural causes are increasing winter air temperatures and precipitation since the 1990s (Fig. 1) [*Iijima et al., 2010; Varlamov et al., 2010*], while the anthropogenic factors include laying underground utility lines, emergency leakage from aqueducts, and disturbance to natural drainage of suprapermafrost groundwaters [*Syromyatnikov and Dorofeev, 2014; Zabolotnik, 2015*].

The geocryological and hydrogeological settings have been monitored at a test site in the southwestern outskirts of Yakutsk, under a low-angle slope of a

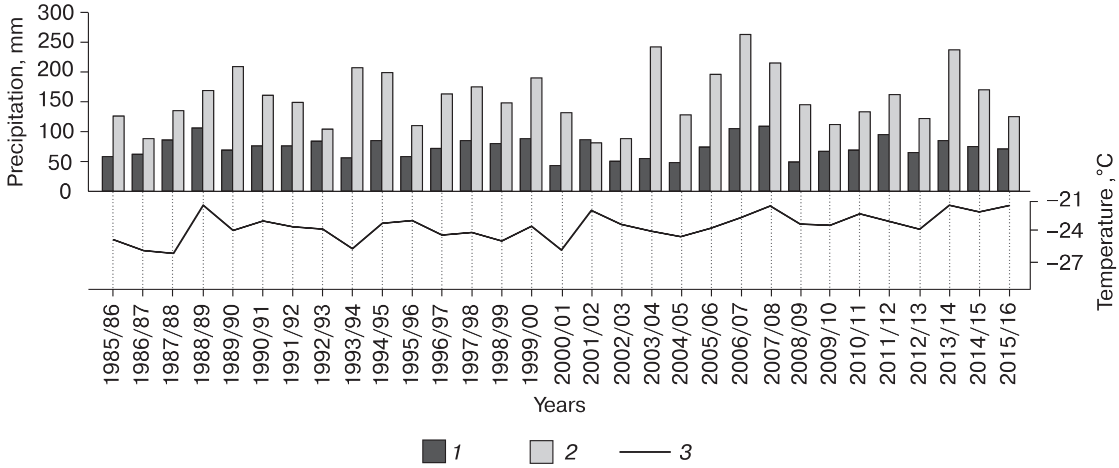

Fig. 1. Variations in total winter (*1*) and summer (*2*) atmospheric precipitation and mean monthly air temperatures (*3*) from October through April in Yakutsk, 1986–2016.

small erosional gully. The territory has been developed since the 1960s. There was a birch wood prior to the construction, and now the gully is grown with bulrush. Quaternary alluvium, 20–25 m thick, is composed of outsize sand lying under 2.0–2.5 m of clay silt and silt. The active layer at the site is 1.8–2.0 m thick, and seasonal temperature variations affect the ground to depths of 16–17 m.

Suprapermafrost cryopegs over an area of 0.06 km^2 were discovered in 1981–1983 when 0.4 to 2.0 m thick moistened sediments were stripped at depths from 1.8 to 3.4 m (interval 1) during exploratory drilling [*Andreev, 1985*]. The groundwater salinity was 4.1 to 12.6 g/L, while the ground temperature was within −0.5 to −0.8 °C. Saline waters found at the active layer base are percolating contaminated waters that drain the surrounding territory and experience cryometamorphism in the conditions of impeded water exchange. Systematic observations of permafrost and chemistry of suprapermafrost waters have been run since 1985. First the monitoring was performed in four holes which were drilled to at least 1 m deep and equipped with filters within the aquifer. In winter 2016, another hole was drilled 1 m far from borehole No. 3. The filters were placed at the depths 1–3 m in borehole 3 and at 4–6 m in the new one. Currently hydrogeological observations are run in both holes.

Intrapermafrost cryopegs stripped within the 7–8 m and 17–19 m depth intervals (intervals 2 and 3, respectively) have been studied since 1988 [*Nim and Fedorov, 1989*]. At the time of drilling, they had a salinity of 26–30 g/L and a temperature of −0.8 to −1.2 °C. The cryopegs of the 7–8 m interval owe their origin to annual and long-term variations in climate parameters [*Anisimova et al., 2001; Anisimova and Pavlova, 2002, 2014*], while those at 10–15 m may result from freezing of relict floodplain and sublake taliks in which non-flowing water becomes highly saline while the ambient rocks are cooling slowly [*Anisimova, 1971*]. Cryopegs at greater depths in alluvial sand within Yakutsk city often form in response to anthropogenic warming of ground containing dissolved salts that migrate from shallower depths. For instance, the ground temperature at the depths 2–7 m became 0.2–0.4 °C higher next to the area where an underground wastewater pipe was laid in 1983. Diffusion redistribution of ions at a high negative temperature led to salinization of alluvial sediments and maintained the formation of a cryopeg at the active layer base (Fig. 2).

The deepest cryopegs have been monitored till present, while observations of the 7–8 m interval stopped in 2004 because the cryopeg had moved [*Pavlova et al., 2009*]. Water level in hydrogeological boreholes was measured every ten days during the first year of observations, and then every month till 2001. However, the sampling strategy changed after main seasonal patterns of the flow regime became clear: water from boreholes was sampled monthly from March to December in the annual cycles of 2015 and 2016 to study variations in the trace-element composition of cryopegs. Temperature-depth profiles were measured in specially drilled geothermal boreholes, 5 to 20 m deep, where the temperature was logged by movable thermistor strings at every 0.5 m above 5 m and at every 1 m below this depth. The ground temperature and water parameters were sampled synchronously.

Additional 5–20 m deep boreholes were drilled and cored in different years to monitor the permafrost and groundwater conditions and to study depth-dependent salinity variations. Water samples and supernatants were analyzed in laboratory by titration and

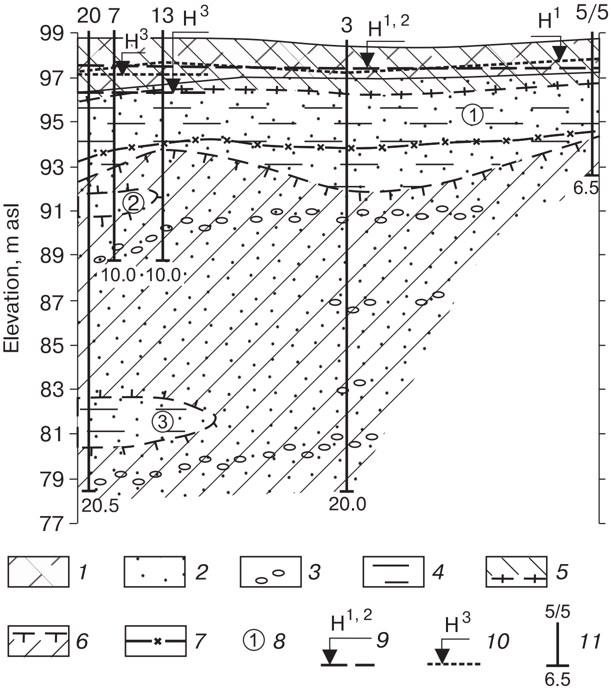

Fig. 2. Generalized cross section of the test site.

1 – alternating silt and clay silt; *2* – outsize sand; *3* – pebble; *4* – water-saturated soil; *5* – active layer and its boundary; *6* – permafrost and its boundary in May 2016; *7* – base of suprapermafrost cryopegs in May 1992; *8* – cryopeg interval; *9*, *10* – hydraulic head of cryopegs in May 1992 (*9*) and May 2016 (*10*), numerals above are cryopeg interval numbers; *11* – hydrogeological borehole: numerals above and below are borehole number and depth (m), respectively.

capillary electrophoresis at the Institute of Permafrost (Yakutsk). In 2015–2016, water chemistry was analyzed by atomic mass spectrometry (AMS) and atomic emission spectrometry with inductively coupled plasma (ICP–AES) at the Analytical Certification and Testing Center of the Institute of Microelectronics Technology and High Purity Materials (IMTHPM, Chernogolovka). REE abundances in cryopegs were normalized to the standard compositions of North American Shale Composite (NASC) and clay from the Russian Platform [*Gromet et al., 1984; Dubinin, 2006*].

RESULTS

Suprapermafrost cryopegs widespread over the territory (Fig. 2) show prominent seasonal and long-term variations in fluid dynamics and water chemistry due to their shallow depths. The aquifer is the thickest (3–5 m) in fall, when its top lies at a depth of 1.8–2.5 m, the waters have the lowest salinity and pH from 7.4 to 9.1 in different parts of the site. The level and salinity of these cryopegs decrease slightly as a result of moisture and salt transport to the region of lower negative temperatures in October, at the onset of silt and clay silt freezing.

The fine-grained sediments over cryopegs freeze up completely in February–March. Since that time, the residual pore water becomes progressively more saline, mainly at the account of Na chlorides and sulfates which have low freezing temperatures, while pH decreases to 6.0–7.3 during pore ice formation in sand. Cryogenic pressure in saline taliks reaches its maximum in May and June (Fig. 3). Thawing of subaerial clay silt begins in the latest May and lasts till mid-July or sometimes as long as September. The water level in the boreholes that strip suprapermafrost cryopegs gradually becomes lower and the salinity decreases due to dilution with molten ground ice. As the ground is thawing, groundwaters gain more hydrocarbonate salts. The water level in the boreholes rises in fall time when cryopegs mix up with percolating meteoric waters; the seasonal water level difference reaches 1.2–2.2 m. The water regime is similar in cryopegs from interval 2, which are thus in close hydraulic connectivity with shallower suprapermafrost groundwaters.

The level and chemistry variations of groundwater depend on weather conditions in specific years, ambient ground salinity, and on the thickness and salinity of cryopegs. The observation period of 1985 to 2016 can be divided into three subperiods according to climate and water chemistry.

1985–1992: lower air temperatures and smaller annual precipitation than in the following years (Fig. 1). In October 1985, suprapermafrost cryopegs with a salinity of 12–14 g/L and a temperature of −0.9 °C were located within a depth interval of 2.2–4.5 m. In the following winter, frost depth reached 3.5 m. In the summer seasons of 1986–1989, sediments above cryopegs locally remained frozen, which favored rapid depthward propagation of the cold front in winter. As the aquifer cooled down to −1.2 °C and the pore water salinity increased to 16–18 g/L, a more saline cryopeg (25–28 g/L) formed at a depth of 7–8 m (interval 2). The relative percentages of salts at that time were: 60 to 66 % Na chlorides, 8 to 15 % Mg chlorides, and 20 to 27 % Mg sulfates (Fig. 4).

1993–2006: slightly warmer winter air temperatures [*Skachkov, 2012*]. During seasonal thawing, the top of the upper aquifer moved deeper (2.5–3.0 m). The ground temperatures around and below cryopegs were −0.8 to −1.1 and −1.2 to −1.4 °C, respectively. Salinity was 15–18 g/L in winter and 10–12 g/L in fall and varied generally from 16 to 22 g/L during the year. Groundwater chemistry in the shallow aquifers consisted mainly of Na chlorides (62–66 %) and Mg sulfates (20–30 %), while Mg chlorides (7–13 %) appeared only in winter; hydrocarbonates and sulfates were within 4–9 % in total.

2006–2016: marked warming of air temperatures and higher precipitation in the beginning of the time

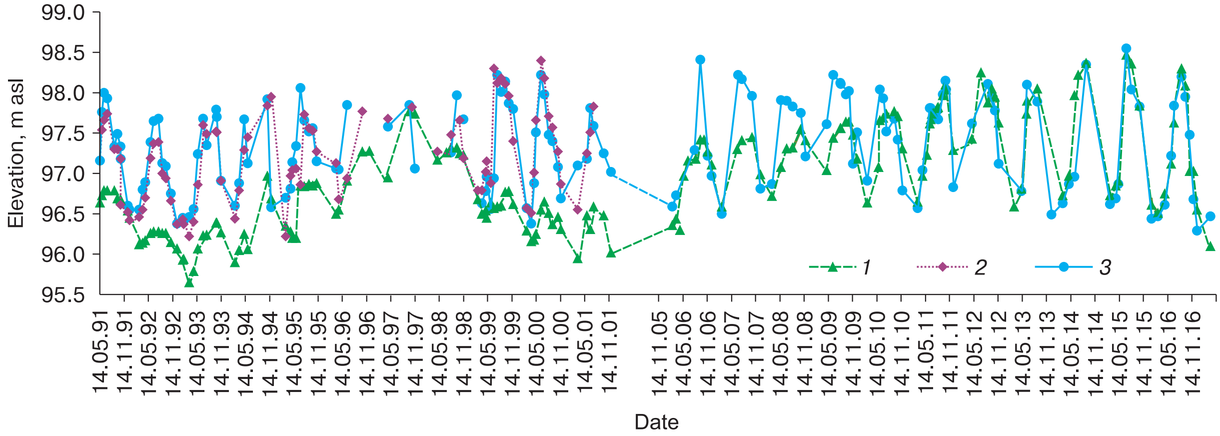

Fig. 3. Long-term variations in water level measured in boreholes at different cryopeg depths.
1 – interval 1 (borehole 3); *2* – interval 2 (borehole 7); *3* – interval 3 (borehole 20).

span. Cryopegs did not penetrate deeper and their salinity did not increase in winter because the winters were relatively mild, with large snow depth and a frost depth no more than 2–3 m. Ground temperature was within –0.5 °C at the cryopeg depths and –0.7 to –0.9 °C underneath.

Groundwaters at the test site have become less saline for the past decade. The salinity of suprapermafrost cryopegs was 7–10 g/L in winter and 3–5 g/L in summer and fall. Although the general salinity decreased, the water composition was still dominated by Na chlorides (53–66 %) and Mg sulfates (20–31 %). Note the presence of Na sulfates (10–13 %) but the absence of Mg chlorides which are indictors of water metamorphism. Similar dilution of suprapermafrost waters was also reported from another site in Yakutsk [*Danzanova, 2014*].

Intrapermafrost cryopegs occur below the active layer (interval 3), and are of local occurrence, according to geophysical and drilling data of different years [*Nim and Fedorov, 1989; Pavlova et al., 2009*]. Compared to suprapermafrost groundwaters, they have similar chemistry but a higher salinity, with its peaks shifted in time.

In the end of the warm season (August–September), intrapermafrost cryopegs are the most saline

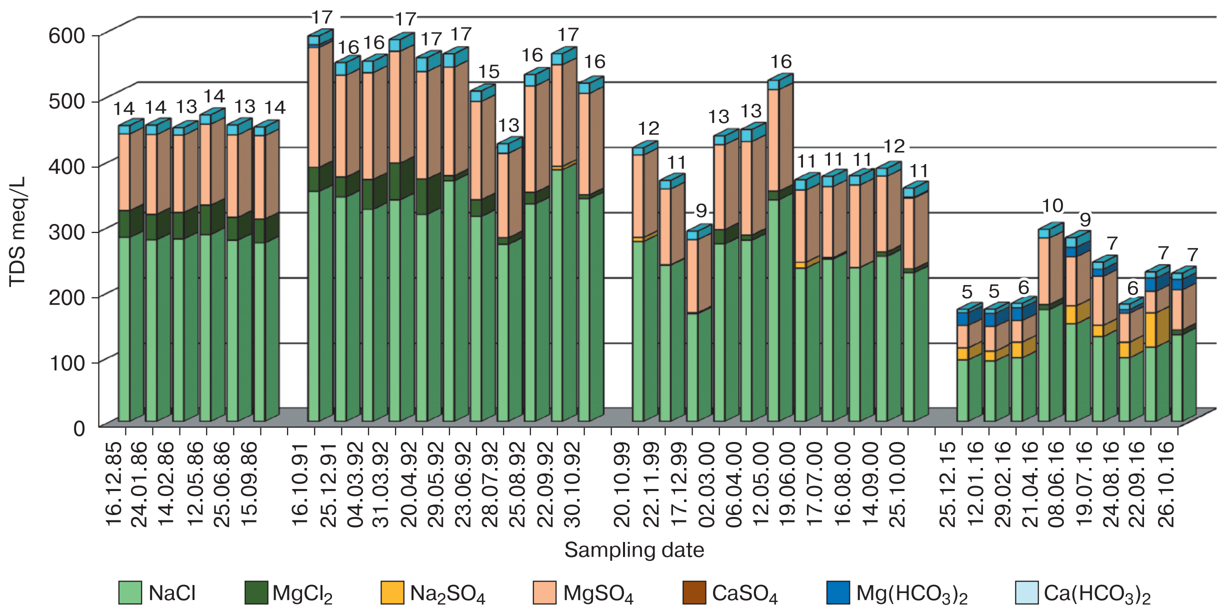

Fig. 4. Long-term variations in salt composition of suprapermafrost cryopegs (borehole 3).
Numerals above the columns are total values, in g/L.

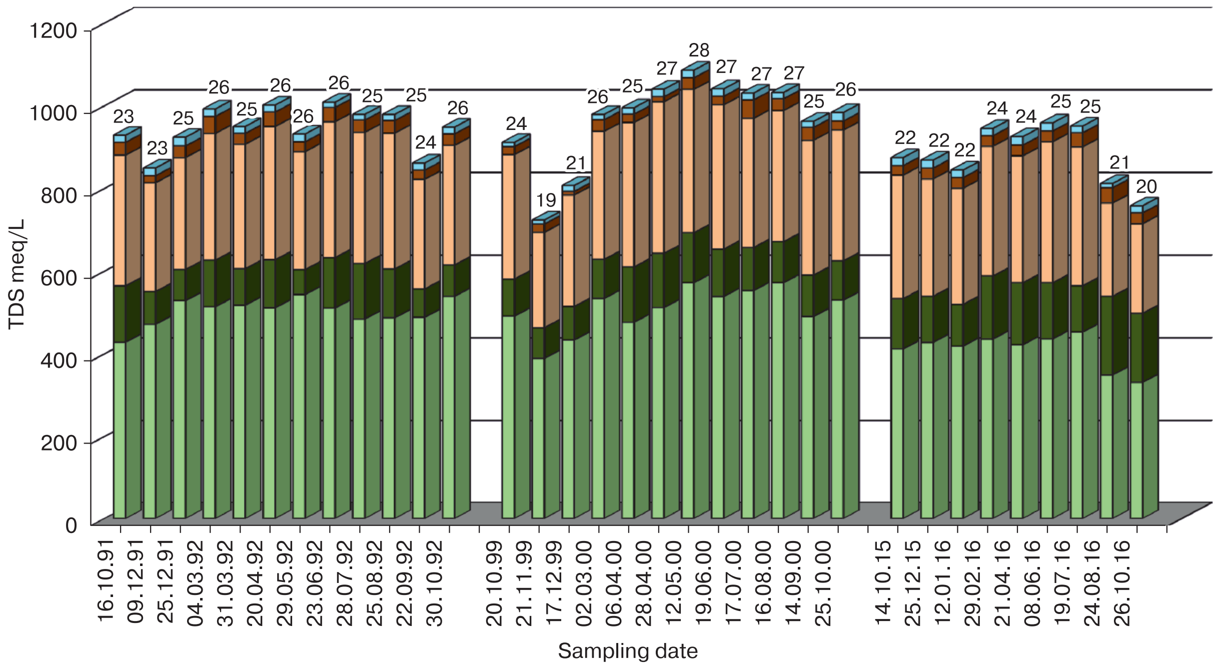

Fig. 5. Long-term variations in salt composition of intrapermafrost cryopegs (borehole 20).

Legend same as in Fig. 4.

(25–30 g/L) and their hydraulic head approaches the ground surface (Fig. 5). From November to March, the hydraulic head becomes deeper and the salinity decreases to 22–26 g/L. Between March and August, the head rises and the salinity increases. The relative percentages of different salts remain invariable for the whole observation period: 55–67 % Na chlorides, 28–35 % Mg sulfates, and 10–14 % Mg chlorides; pH within 6.3–7.3. The ground temperature is stable at −1.1 to −1.2 °C.

The long-term cycle comprises two subperiods that differ in fluid dynamics and do not coincide with the subperiods distinguished for suprapermafrost groundwaters.

1988–2009: lower water level than in the overlying aquifer (Fig. 2), the difference being the greatest in May–July (1.2–1.6 m) and the smallest in November–December (0.2–0.6 m).

2009–2016: progressive leveling of suprapermafrost and intrapermafrost cryopegs between 2009 and 2011 when the water level became almost the same in all boreholes, which indicates improved hydraulic connectivity of groundwaters circulating at different depths (Fig. 3). The fluid dynamic settings may have changed in response to weather changes of 2006–2008. In those years, precipitation increased all over Central Yakutia and provided significant inputs to both suprapermafrost and intrapermafrost groundwaters. The enhanced connectivity among aquifers is confirmed by salinity decrease (to 18–25 g/L) in the deepest cryopegs in 2015–2016.

In 2015–2016, first data were obtained on the contents and variations of trace elements (including REE) in cryopegs from the test site (Table 1). The suprapermafrost cryopegs have 3–5 times lower abundances but more diverse and variable compositions of trace elements than the intrapermafrost cryopegs. The suprapermafrost groundwaters contain Al, Si, Ba, Mo, Sn, Sb, Ce, Hf, Th, and U, while the intrapermafrost cryopegs have greater enrichment in mobile elements (F, Li, Sr, Br). This depth-dependent differentiation may be due to high clay contents in shallower sediments, as well as to lower ground temperatures at 17–19 m which limit the leaching capacity of cryopegs.

The contents of most trace elements in cryopegs do not show notable seasonal variability, with some exceptions. Namely, Li, Sr, and Zn are the highest in the end of the freezing season when the water becomes more saline and has a lower pH, while Mo, Sn, and Sb are slightly higher in fall.

REE contents in the sampled cryopegs vary notably over the year. They are commonly low in summer but higher in winter when the ground is frozen. They are mostly (82–96 %) La to Sm LREE. The REE profiles of cryopegs normalized to the contents in the NASC and Russian Platform clay standards are similar and relatively smooth.

Table 1. **Element contents in cryopegs**

Element		Borehole number and cryopeg depth			
		Borehole 3 (1.8–6.0 m)	Borehole 5/5 (1.5–3.0 m)	Borehole 13 (1.8–4.2 m)	Borehole 20 (16.0–18.0 m)
		min–max / average	min–max / average	min–max / average	min–max / average
pH		6.6–8.5 / 7.6	6.8–9.8 / 8.0	6.0–9.7 / 7.7	6.1–7.9 / 7.1
F	mg/L	0.5–3.1 / 1.6	0.4–2.3 / 1.1	0.3–3.9 / 1.2	0.2–12.7 / 3.4
Br	mg/L	1.8–5.7 / 3.3	1.5–4.8 / 3.1	2.2–7.1 / 4.4	13.2–19.2 / 15.9
Ba	mg/L	0.02–0.2 / 0.1	0.007–0.1 / 0.07	0.01–0.4 / 0.2	0.02–0.05 / 0.04
Sr	mg/L	0.6–2.6 / 1.5	0.4–4.0 / 1.8	1.1–3.9 / 2.5	11.4–15.1 / 12.8
Al	mg/L	0.04–4.8 / 2.3	0.07–4.7 / 1.5	0.07–67.2 / 8.3	0.07–0.9 / 0.3
Fe	mg/L	0.6–115.6 / 40	0.3–126.0 / 52	0.9–374.0 / 134	17.0–174.0 / 67
Mn	mg/L	0.3–4.0 / 1.3	0.01–2.7 / 1.2	1.0–13.9 / 2.5	1.4–2.7 / 2.0
Si	mg/L	0.6–5.9 / 3.4	0.4–5.9 / 3.2	0.3–25.2 / 6.8	0.3–1.4 / 0.7
Zn	mg/L	0.007–0.2 / 0.07	0.008–0.5 / 0.2	0.07–2.6 / 0.7	0.02–0.3 / 0.1
Li	μg/L	2.1–11.1 / 6.4	1.1–9.3 / 5.3	2.8–16.7 / 13.9	15.6–18.9 / 17.5
Cs	μg/L	0.01–0.2 / 0.12	0.01–0.1 / 0.08	0.02–0.7 / 0.2	0.05–1.0 / 0.07
Mo	μg/L	1.5–20.7 / 12.5	0.8–28.7 / 9.2	0.5–17.2 / 6.8	1.2–6.8 / 3.7
Sn	μg/L	0.3–24.7 / 10.7	0.3–18.8 / 7.0	0.4–5.4 / 2.0	1.5–5.3 / 2.8
Sb	μg/L	0.3–1.0 / 0.6	0.2–1.0 / 0.6	0.3–1.6 / 0.9	0.3–1.0 / 0.6
Hf	μg/L	0.03–0.4 / 0.2	0.02–0.3 / 0.2	0.07–1.4 / 0.4	0.07–0.2 / 0.1
Th	μg/L	0.03–9.0 / 3.7	0.07–7.5 / 2.4	0.09–41.9 / 7.6	0.08–1.3 / 0.4
U	μg/L	4.5–90.2 / 49.6	11.6–150.1 / 47.9	6.5–224.1 / 92.5	0.3–1.7 / 0.9
Σ REE	%	0.8–242 / 91	0.7–145 / 53	1.2–2198 / 334	1.5–97 / 8.0
Σ LHREE	%	82–92 / 86	84–94 / 86	84–87 / 85	85–96 / 89
La_n/Yb_n		2.2–3.5 / 2.9	2.2–3.6 / 2.9	2.6–3.2 / 2.9	0.8–6.0 / 2.5

N o t e: numerator and denominator show, respectively, variation ranges and average values. La_n/Yb_n ratios are normalized to NASC standard [*Gromet et al., 1984*]. F contents and pH were determined by potentiometry at the Institute of Permafrost (Yakutsk); other elements were analyzed by AMS and ICP-AES at the Analytical Certification and Testing Center of the Institute of Microelectronics Technology and High Purity Materials (Chernogolovka).

REE are the lowest in intrapermafrost cryopegs which occur in almost clay-free sediments. Judging by REE contents compared in filtered and non-filtered waters, the elements are carried on particulate matter. REE in suprapermafrost waters become 4–6 times lower upon filtration (0.45 μm cellulose filter), though the reduction for intrapermafrost waters is by a factor of 0.5–0.8 only.

The contents of REE in cryopegs poorly correlate with most of major elements and with pH. However, there is some correlation with Fe and Ca, to $r = 0.6$ and 0.7, respectively, and a high correlation with Al, Si, and Mn ($r = 0.98$, 0.95 and 0.96, respectively). The REE correlation with Al, Si, and Mn may result from REE inputs during weathering and leaching of aluminosilicate host rocks and active involvement of the elements into formation of secondary phases.

CONCLUSIONS

Years of observations at the test site within Yakutsk city have demonstrated that the fluid dynamics and water chemistry of suprapermafrost cryopegs are controlled jointly by air temperatures and atmospheric precipitation. Several successive cold and low-snow winters can lead to rapid depthward propagation of the freezing front while cryopegs move to greater depths and residual unfrozen pore waters become more saline. On the other hand, aquifers become thicker while the salinity of suprapermafrost groundwaters decreases after relatively mild winters with large snow depth.

Cryopegs that occur at different depths share similarity in water chemistry and show synchronicity in water level variations, which indicates hydraulic connectivity within the active layer. Improved connectivity under the recent climate warming can be inferred from water level similarity in all boreholes and moderate dilution of deeper cryopegs following that of suprapermafrost waters.

Suprapermafrost groundwaters show greater diversity of trace-element compositions, but intrapermafrost cryopegs have higher enrichment in more mobile F, Li, Sr, and Br, which can accumulate in the conditions of impeded water exchange.

If the current ground warming continues, the concentrations of trace elements, including toxic components, can be expected to increase in pore waters of shallow aquifers, where the physicochemical water-rock interaction is especially active.

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