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FLUORIDE DISTRIBUTION IN SUBPERMAFROST GROUNDWATER IN CENTRAL YAKUTIA

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We studied the distribution of fluoride (F^-) in fresh and moderately mineralized waters of subpermafrost aquifers in Central Yakutia. We analyzed the data archive of 296 water samples collected by the Melnikov Permafrost Institute staff members during hydrochemical surveys between 1984 and 2019. The average fluoride concentrations varied between 5 and 10 mg/L. Highest concentrations (up to 15.5 mg/L) were determined in waters of the terrigenous aquifers over the crystalline basement. The sources of fluoride can likely be the various fluorine-bearing minerals of aluminosilicate rocks. The high fluoride concentrations in these aquifers are associated with geochemistry (alkaline medium and sodium bicarbonate water type) resulting from cryogenic metamorphism of rocks. Lowest concentrations (0.4–0.8 to 2–3 mg/L) were found in subpermafrost water samples collected from boreholes near the Lena River channel. These low concentrations indirectly indicate the presence of open taliks under the channel and the infiltration of stream water into the subpermafrost aquifers.

Keywords: *artesian basin of Yakutia, subpermafrost water, permafrost, low groundwater circulation zone, chemical composition, fluoride*

INTRODUCTION

Surface waters are the main source of fresh drinking water in the continuous permafrost area of Central Yakutia. Waters of hydrogenous taliks, as well as subpermafrost waters are less commonly used. Concentration of fluoride is one of the water quality indicators. The optimal concentration lies within the range of 0.5–1.0 mg/L, while the maximum allowable concentration cannot exceed 1.2–1.5 mg/L [*Fordyce et al., 2007; Ozsvath, 2009; Donskikh, 2013; WHO, 2017; Yousefi et al., 2019*]. Long-term consumption of water with high fluoride concentration (>1.5 mg/L) has a toxic effect on the musculoskeletal, neuroendocrine, and cardiovascular systems, while the low concentrations (<0.5–0.6 mg/L) lead to the development of dental caries [*Brindha and Elango, 2011; Donskikh, 2013*].

Fluorine enters water solutions from rocks, in which it is normally present as a component of sellaite (MgF_2), fluorite or fluorspar (CaF_2), cryolite (Na_3AlF_6), and fluorapatite ($Ca_5(PO_4)_2F$) minerals. The fluorine abundance in the lithosphere is estimated at 400–800 mg/kg [*Vernadsky, 1955; Yanin, 2007*]. The highest content of fluorine is in granites: 500 to 1400 mg/kg [*Krauskopf and Bird, 1995*], 810 mg/kg on the average [*Wedepohl, 1969*]. Its content reaches 330 mg/kg in calcareous rocks, sands, and sandstones and is up to 610 mg/kg in clays and shale [*Grigoriev, 2009*]. The concentration of fluoride in ocean water is approximately 1.3 mg/L [*Turekian, 1972; Bordovsky and Ivanenkov, 1979; Gordeev, 1983*]. The range of fluoride concentrations is wide in fresh and slightly saline natural waters: from trace amounts in river water to 16 mg/L and higher in underground aquifers [*Gordeev, 1981; Kraynov and Shvets, 1987; Shvartsev et al., 2007; Anichkina, 2016*]. Normal fluoride concentrations in natural waters are often exceeded in tropical climate [*Shvartsev et al, 2007; Brindha and Elango, 2011; Subba Rao, 2017; Malago et al., 2017*]. Evaporative concentration, ion exchange, and technogenic groundwater pollution by wastewaters from various sources (chemical, rubber, electrical industry, non-ferrous and ferrous plants, mechanical engineering, as well as coal-fired power plants) are the main processes that lead to the formation of fluoride-rich waters) [*Kraynov and Shvets, 1987; Shvartsev et al., 2007; Yanin 2007*].

Data on fluoride concentrations in fresh groundwaters in permafrost areas are rather scarce and mainly concern water in the zone of free water exchange. It is known that the fluoride concentration in the groundwater of West Siberian artesian basin lies within sanitary standards [*Beshentsev, 2013*]. The prevailing concentration of 0.2–0.6 mg/L is observed in the spring water of Chita region with isolated permafrost distribution [*Zamana et al., 2011*]. The average fluoride concentration of 0.1–0.3 mg/L is found in supra-permafrost waters of arctic and subarctic regions of Russia, Alaska, and Canada [*Shvartsev, 1978*], while in mountainous fluorine-bearing provinces it can reach 0.7 mg/L [*Kraynov and Shvets, 1987*]. The concentration of fluorine varies from trace amounts to 0.4 mg/L in fresh groundwater of the Aldan Highland (East Siberia) with discontinuous permafrost [*Filimonova, 1977*]. Elevated concentrations of fluoride (up to 2.7 mg/L) have only been documented in water samples extracted from explorational boreholes in river valleys over large tectonic faults.

Earlier, the analysis of fluoride concentrations in subpermafrost groundwater confined to the zone of impeded water exchange in was conducted by N.P. Anisimova and T.M. Golovanova [*Anisimova, 1958, 1981; Anisimova and Golovanova, 1972*]. These works pointed out the significant (one-order magnitude) differences in fluoride concentrations that could be observed within groundwater of the same aquifer. It was also found that the concentration of fluoride in subpermafrost groundwater increases with an increase in waster alkalinity and the concentration of sodium bicarbonates.

At present, there is a large amount of data on fluoride concentrations in groundwater of Central Yakutia, which requires the synthesis and analysis. The aim of this study is probability assessment of finding elevated fluoride concentrations within aquifers of this region.

MATERIALS AND METHODS

Main sources of information for this study were legacy hydrochemical data collected in Central Yakutia by staff members of the Melnikov Permafrost Institute, Siberian Branch of the Russian Academy of Sciences in 1984–2019 and data from repositories of various geological organizations. To characterize the chemistry of subpermafrost aquifers and the concentration of fluoride in them, data on 296 water samples were analyzed. Most of the samples (153 samples) were obtained in the city of Yakutsk and its vicinities. Nowadays, there are about 50 deep water wells providing technical water supply to various enterprises. Water treatment systems including defluorination of subpermafrost water and its use for drinking purposes are installed on several of these drilled wells. Less studied are fresh and slightly saline (<3 g/L) subpermafrost waters of the Lena–Aldan (134 samples) and Lena–Vilyui (nine samples) interfluves. Hydrochemical data from samples of lake and river water (183 samples) were also considered for fluoride concentration assessment in surface waters.

Water samples with the total mineralization of less than 3 g/L were considered while processing the results of chemical analyses. Fluoride concentrations in water were determined by potentiometric method. Before 2000, an electrode system consisting of a fluoride selective electrode and a silver chloride reference electrode. Later, an ion-selective single-crystal lanthanum fluoride membrane electrode was applied.

NATURAL CONDITIONS OF CENTRAL YAKUTIA

Study area is located within the Central Yakutian accumulative plain with absolute heights ranging between 200 and 400 m asl in the middle reaches of the Lena River and its tributaries Aldan and Vilyui. Snowmelt and rainwater are the main sources of river discharge. The contribution of groundwater is only about 13–17 % [*Dzhamalov et al., 2012*]. In rivers, the content of total dissolved solids (TDS) during the summer usually does not exceed 0.05–0.4 g/L; the chemical composition is dominated by bicarbonate and magnesium and calcium ions; the reaction is neutral. In winter, the TDS contents increases up to 0.3–0.7 g/L under the lack of atmospheric precipitation, and the chemical composition turns to chloride-bicarbonate with mixed cations [*Anisimova and Pavlova, 2014*]. In low-water winter conditions, the TDS content in the Vilyui and Aldan rivers is 0.1–0.2 g/L higher compared to that in summer without changes in dominating ions. In winter, small rivers/streams do normally freeze to the bottom. Average fluoride concentrations in these streams do not significantly differ from average concentrations in rivers beyond the permafrost zone and do not reach optimal levels for drinking purposes.

Lakes of various geneses are widespread in the study area. Ionic composition of freshwater lakes is of the bicarbonate calcium–magnesium or magnesium-calcium types [*Anisimova, 1981*]. Slightly saline (1–3 g/L) lake waters are dominated by bicarbonates among anions, and by sodium and magnesium among cations. Water pH varies from neutral to strongly alkaline values; fluoride concentration, from 0.2 to 1.1 mg/L reaching 1.7–2.3 mg/L in closed thermokarst and erosional-thermokarst lakes.

In terms of hydrogeology, the area belongs to the Yakutsk artesian basin, the sedimentary cover of which consists of terrigenous-carbonate deposits of Paleozoic and Mesozoic ages. In general, the sedimentary cover deposits lie gently with an inclination towards the inner part of the basin. Sediment thickness reaches 1–4 km and more decreasing in to 538–890 m the area of Yakutsk arch uplift [*Balobaev et al., 2003*]. The territory is underlain by continuous permafrost with the average thickness of 300–450 m reaching the maximum of 885 m within the Buotama–Amga interfluve [*Balobaev et al., 2003*]. A significant decrease in permafrost thickness and appearance of open taliks can only be observed under the Lena River channel, its main tributaries, and under several lakes.

Under the regional cryogenic aquiclude, there are subpermafrost aquifers with hydrological regimes depending on the long-term climatic changes. Nowadays, permafrost is being thawed from the bottom [*Balobaev, 1991*]. Most of researchers explain the low water pressure and low salinity of subpermafrost waters in the central part of the artesian basin by the uplift of permafrost base [*Kononova, 1973; Romanovskii, 1983; Shepelev et al., 1984; Balobaev et al., 2003*]. The composition of trace elements in subperamfrost waters is relatively stable. Fluorine is one of the constantly occurring elements.

SUBPERMAFROST WATERS AND FLUORIDE DISTRIBUTION

In Central Yakutia, subpermafrost waters occur within the Archean formations of the crystalline basement and aquifers of the sedimentary cover composed of carbonate rocks of the Upper Proterozoic (Vendian) and Cambrian, as well as terrigenous Jurassic and Cretaceous deposits (Fig. 1).

Archean basement rocks consist of biotite gneisses, granite-gneisses, and crystalline schist. These are located at depths of 560–589 m in the central part of Yakutsk arch uplift and at a depth of 1022 m in the Amga trough [*Balobaev et al., 2003*]. The water content in the basement rocks is relatively low. The TDS content in the basement groundwater is 4.0–4.3 g/L; the ionic composition is dominated by sulfate or sulfate–bicarbonate anions and sodium cations. There is no information about the fluoride concentrations in these waters, except for a single borehole drilled on the floodplain of the Lena River. In this borehole, the concentration of fluoride in groundwater of Archean rocks is 2.2 mg/L. However, this value can possibly be not representative, because the borehole is close to the open talik under the Lena River channel. Northward, in the area of the Yakutsk arch uplift, the concentration of fluoride in groundwater of Archean and Lower Jurassic aquifers reaches 8.8–9.0 mg/L.

The Vendian and Lower Cambrian rocks are represented by fractured dolomites and dolomitic limestones bituminous to varying degrees. These rocks were studied in the southern part of Central Yakutia up to the latitude of Yakutsk. They are located at depths ranging from 285 to 520 m. The TDS content in the groundwater of these aquifers is 3.5–4.4 g/L; in the chemical composition, sulfate or bicarbonate–chloride anions and sodium cations predominate; less often, there is mixed cation composition; the reaction is neutral or slightly alkaline; fluoride concentrations vary from 1.7 to 4.4 mg/L.

The Middle Cambrian deposits are widespread within the northern flank of the Aldan anteclise, except for the Yakutsk uplift, where Jurassic deposits overlie Archean formations [*Balobaev et al., 2003; Shepelev and Makogonova, 2010*].

The Middle Cambrian aquifer is composed of limestones, marls, and dolomites. It has been found while drilling at depths ranging from 180 to 422 m. The TDS content in the groundwater of this aquifer is within 0.9–1.9 g/L. The fluoride concentration in bicarbonate and chloride–bicarbonate water with neutral or slightly reaction varies within 3.4–8.8 mg/L (Figs. 2 and 3). Less often, the Middle Cambrian deposits contain bicarbonate–sulfate water with mixed composition of cations, neutral reaction, and fluoride concentration of 1.3–2.2 mg/L.

In Yakutsk, aquifers associated with terrigenous Jurassic and Cretaceous rocks have been studies rela-

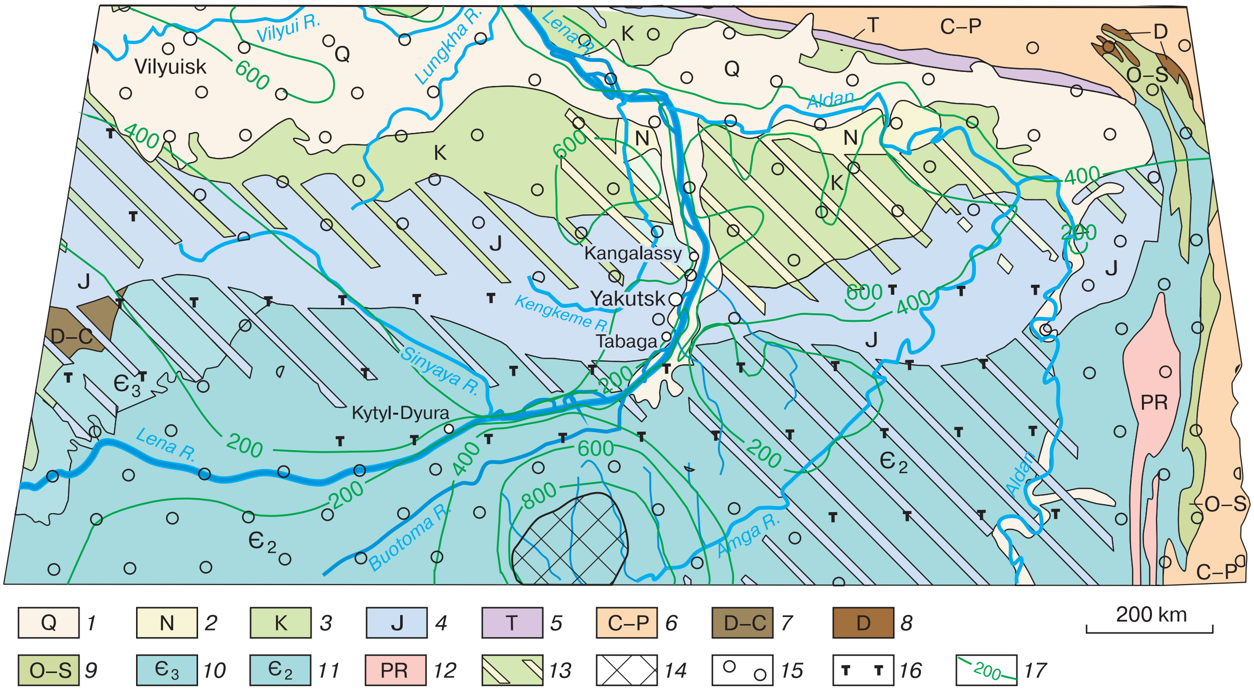

Fig. 1. Schematic hydrogeological map of the study area (modified after [*Efimova and Zaitsev, 1970*; *Melnikov and Tolstikhin, 1983; Balobaev et al., 2003; Semenov and Zheleznyak, 2018*].

Aquifers within: (*1*) Quaternary, (*2*) Neogene, (*3*) Cretaceous, (*4*) Jurassic, (*5*) Triassic, (*6*) Carboniferous and Permian, (*7*) Devonian and Carboniferous, (*8*) Devonian, (*9*) Ordovician and Silurian, (*10*) Upper Cambrian, (*11*) Lower Cambrian, and (*12*) Upper Proterozoic deposits; (*13, 14*) sediment complexes that do not contain gravity water in the liquid phase: (*13*) frozen aquifers (the color of narrow bands corresponds to the age of the frozen complex, and the background color corresponds to the age of the first aquifer from the surface), (*14*) absence of subpermafrost water in the basin's sedimentary cover; TDS content: (*15*) <1 g/L, (*16*) 1–3 g/L; (*17*) isolines of the thickness of the cryogenic aquiclude.

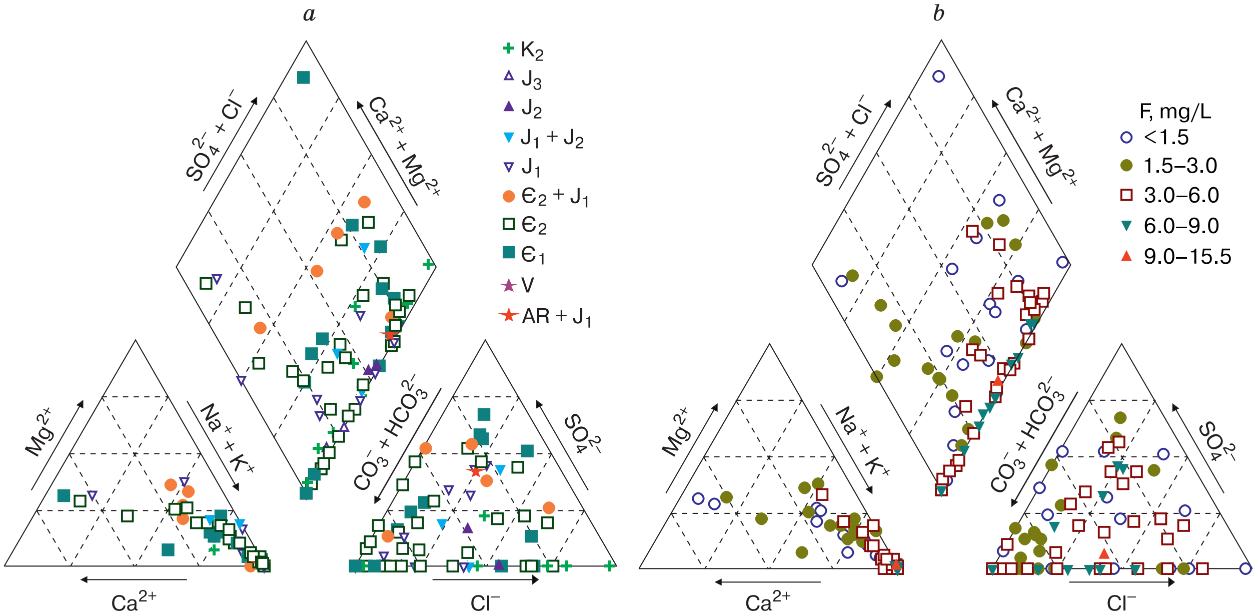

Fig. 2. Piper diagrams of the macrocomponent composition of groundwater (*a*) and fluoride concentrations (*b*) in different aquifers of the Lena–Aldan interfluve.

K_2 – Lower Cretaceous aquifer, J_3 – Upper Jurassic aquifer, J_2 – Middle Jurassic aquifer, $J_1 + J_2$ – jointly sampled Middle and Lower Jurassic aquifers, J_1 – Lower Jurassic aquifer, Є$_2$ + J_1 – jointly sampled Lower Jurassic and Middle Cambrian aquifers, Є$_2$ – Middle Cambrian aquifer, Є$_1$ – Lower Cambrian aquifer, V – Vendian aquifer, AR + J_1 – jointly sampled Lower Jurassic and Archean aquifers.

tively well [*Balobaev et al., 2003; Shepelev and Makogonova, 2010; Pavlova and Fedorova, 2020*]. Scarce data on these aquifers are available for the Lena–Amga and Lena–Vilyui interfluves. At the base of the Lower Jurassic section, there are basal conglomerates overlain by quartz and feldspar–quartz sandstones with siliceous and ferruginous cement. This layer is overlain by marine deposits (sandstones interbedded

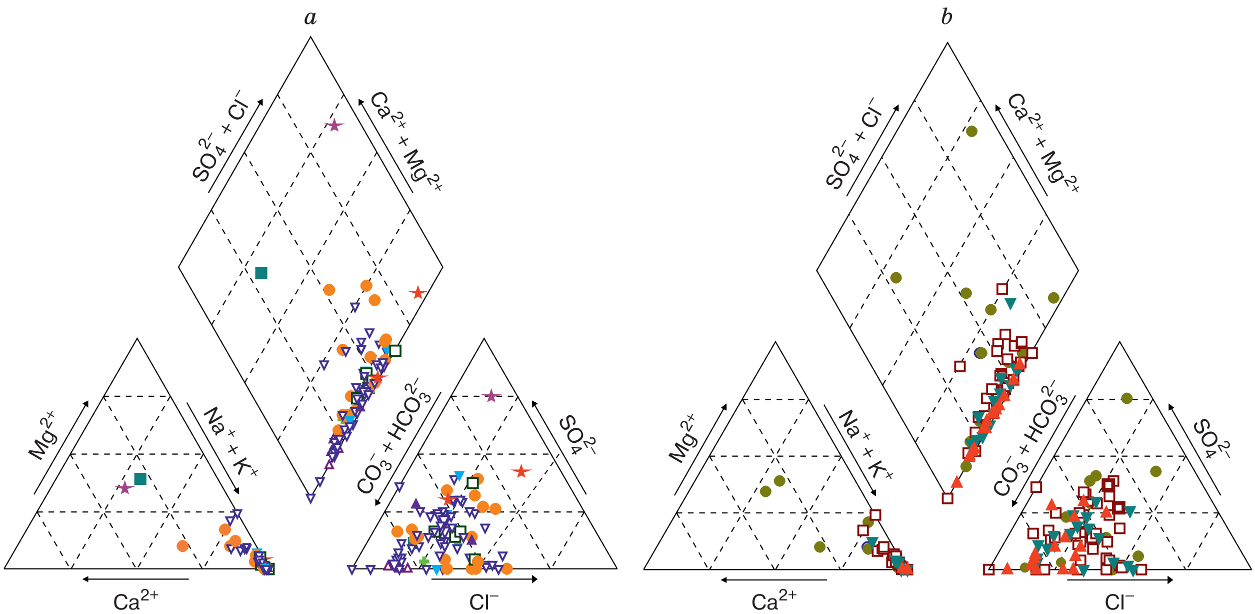

Fig. 3. Piper diagrams of the macrocomponent composition of groundwater (*a*) and fluoride concentrations (*b*) in different aquifers of the Lena–Vilyui interfluve.

For legend, see capture to Fig. 2.

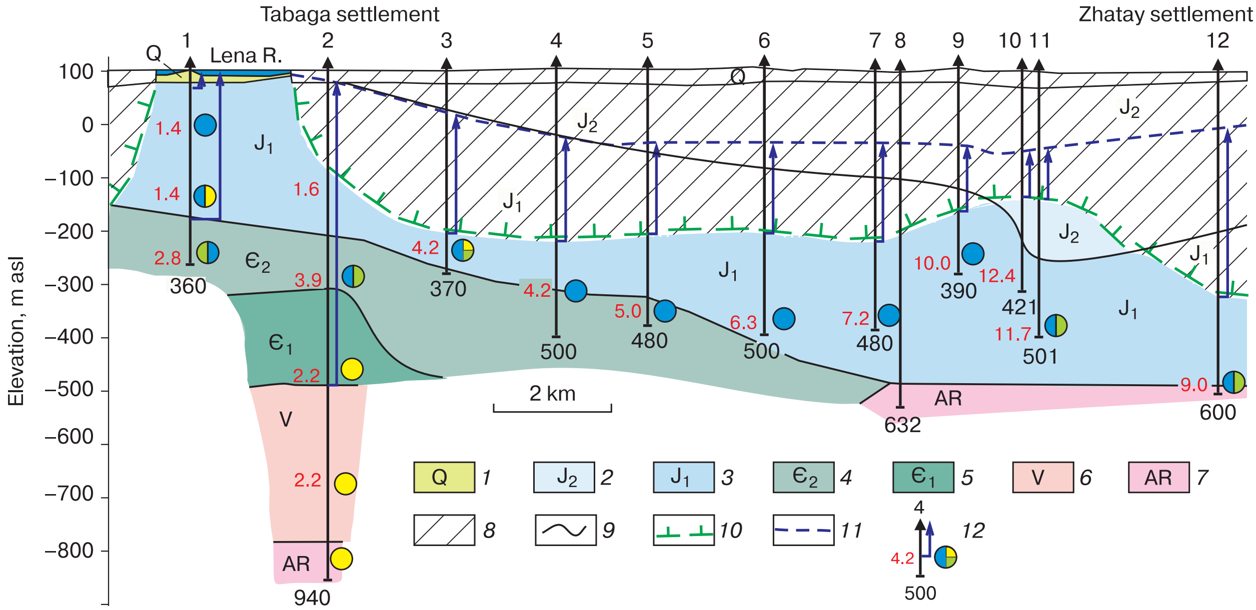

Fig. 4. Hydrogeological section along the line between Tabaga and Zhatay settlements (drawn according to legacy data of the Melnikov Permafrost Institute, Siberian Branch of the Russian Academy of Sciences).

Aquifers: (*1*) Quaternary alluvial, (*2*) Middle Jurassic terrigenous, (*3*) Lower Jurassic terrigenous, (*4*) Middle Cambrian carbonate, (*5*) Lower Cambrian carbonate, (*6*) Upper Proterozoic terrigenous-carbonate, (*7*) locally aquiferous fractured zone of the Archean crystalline basement; (*8*) cryogenic aquiclude; (*9*) boundaries between deposits of different ages; (*10*) permafrost boundary; (*11*) piezometric level of subpermafrost water; (*12*) hydrogeological borehole (figure at the top is its id number, figure at the bottom is the borehole depth, m; on the left, fluoride concentration in water (mg/L) is indicated). The color corresponds to the dominating anions in the water: blue – bicarbonate, yellow – sulfate, green – chloride. Arrows indicate groundwater pressure.

with siltstones and rare lenses of limestones and shell rocks). In the Lena–Amga interfluve, the Lower Jurassic aquifer lies at a depth of 563 m, while in the

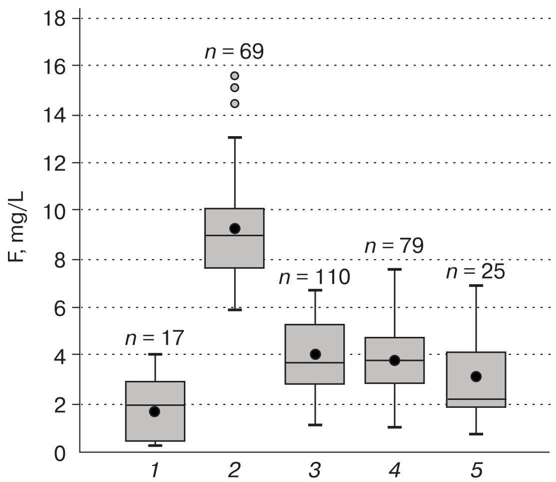

Fig. 5. Boxplots of fluoride concentration in subpermafrost water of Central Yakutia (*n* – sample size).

1 – Cretaceous and Upper Jurassic aquifers; *2*, *3* – Lower Jurassic aquifer complex (*2* – over crystalline basement rocks in the area of the Yakutsk arch, *3* – over Cambrian deposits); *4* – jointly sampled Lower Jurassic and Middle Cambrian aquifers; *5* – Middle Cambrian aquifer.

area of Yakutsk, at 173–420 m. In a larger part of the city, the subpermafrost water of this aquifer is hydraulically connected with the Middle Cambrian aquifer; together, they represent a single pressure system of formation water in pores and fractures [*Balobaev et al., 2003*].

Water-saturated Middle Jurassic formations were studied in the north of Yakutsk. These formations consist of sands interbedded with siltstones, silt, and quartz fine-grained sandstones; clays and shales are often found in the section. The top of this aquifer lies at a depth of 380–500 m [*Shepelev and Makogonova, 2010*]. The Upper Jurassic aquifer is found to the north of Kangalass Promontory latitude . It consists of continental formations (sands and sandstones with interlayers and lenses of coal) at depths of 200–540 m. It's top descends to depths 1800–2496 m within the Lungkhinskaya and Nizhnealdanskaya depressions, where the Lower Cretaceous subpermafrost aquifer composed of interbedded sands, sandstones, mudstones, and clays is the closest to the surface. In the Nizhnealdanskaya depression, water-saturated Lower Cretaceous deposits lie under the permafrost with a thickness varying from 270 to 415–610 m.

In general, the concentration of fluoride varies within the range of 4.1–11.7 mg/L in subpermafrost waters confined to the Lower and Middle Jurassic deposits. Local rise in its concentration (9.0–15.5 mg/L) is observed in the area of Yakutsk arched uplift (Fig. 4). This uplift can be traced in a series of boreholes on the left bank of the Lena River in the northern part of Yakutsk (Lake Beloe, Zhatay, and Markha settlements), and on the right bank in the Tekhturskaya borehole (in the mouth of the Suola River). Here, Cretaceous deposits overlie the Archean rocks (depths ~560–600 m), whereas Cambrian aquifers are absent. The TDS content in water of Jurassic aquifer is 0.9–1.5 g/L; sodium bicarbonates predominate (Figs. 2 and 3). This water has a somewhat increased alkalinity (slightly to moderately alkaline reaction). The proportion of calcium, which could form secondary minerals with fluorine, can rarely reach 2 % (mg-eq/L).

Subpermafrost groundwater of the slightly lithified Cretaceous and Upper Jurassic rocks has the TDS content of 0.5–1.2 g/L; it is of the sodium chloride–bicarbonate or bicarbonate composition and has the fluoride concentration of 1.6–4 mg/L.

DISCUSSION

Fluoride concentration in subpermafrost water of Central Yakutia varies in the range of several tenths to 15 mg/L. There is no evident direct relationship between the fluoride concentration and the aquifer depth, as well as the modern thickness of overlying cryogenic aquiclude. However, the influence of long-term freezing on the accumulation of fluoride in the subpermafrost groundwater cannot be excluded. It is known that the dissolution of fluorine minerals increases under alkaline conditions in water with the TDS content of 0.7–1.7 g/L [*Saxena and Ahmed, 2001*]. Cryogenic transformation of subpermafrost water in Central Yakutia–precipitation of difficultly soluble calcium carbonates during freezing of aquifers, desulfatization (sulfate reduction) of water under anaerobic conditions in the presence of organic matter – is the main control of increased alkalinity and low calcium concentration [*Kononova, 1973; Anisimova, 1981; Fotiev, 2009*]. The desulfatization process is possible under both degradation and aggradation of permafrost [*Anisimova, 1981*]. The fluoride source is the aquifer rock. The concentration of fluoride in subpermafrost water considerably increases under conditions of impeded water exchange and changes in the base of the cryogenic aquiclude within the Yakut arch uplift, where terrigenous sandstones of the Lower Jurassic aquifer are in contact with fluorine-rich granitic rocks (Fig. 5). According to V.T. Balobaev [*1991*], the modern (~10 000 years) uplift rate of permafrost base around Yakutsk can be estimated at 1.7 cm/yr and is due to lower values of heat flux in the frozen zone compared to the thawed subpermafrost zone. While permafrost is degrading, alkaline confined aquifer water migrates into the thawed rock layers and mixes with the low-mineralized water [*Kononova, 1973; Anisimova, 1981*]. As a result of the interaction between the desalinated solution and fluorine-containing minerals, the fluoride transfer from rocks and its concentration in subpermafrost water is possible.

According to [*Shvartsev, 2017*], fluoride concentration directly reflects the time of evolution of the water–rock system: the longer this time, the higher the fluoride concentration. Anomalously low hydrostatic pressure in subpermafrost complexes attests to the long interaction between subpermafrost water and host rock and to significantly worse water exchange conditions in the central and northern parts of the study area. In the area of the Yakutsk arch uplift and the Nizhnealdanskaya depression, the piezometric level of groundwater is close close to zero or even below sea level. Such hydrodynamic situation is formed under conditions of continuous permafrost degradation, when the emerging due to phase transitions free volume is not compensated by the water inflow.

The relatively low concentration of fluoride is characteristic for the subpermafrost water within the right bank of Lena River to the south of Yakutsk latitude. The geological and hydrogeological structure of this territory has the following features: (1) intrapermafrost aquifers (taliks) with fresh groundwater depleted in fluoride are widely distributed within the sandy deposits of medium-altitude terraces of the Lena River; (2) there are open taliks under large lakes that serve as pathways between surface, intrapermafrost and subpermafrost waters; (3) there is a reduction in the thickness of the cryogenic aquiclude to 60–100 m in areas disturbed by tectonic faults, and both vertical and lateral water flows take place along systems of cracks and karst cavities in calcareous rocks; (4) up to the latitude of Tabaga Promontory, free water outflow from boreholes penetrating subpermafrost aquifers takes place. These four controls determine the possibility of subpermafrost water exchange with the surface and atmospheric water and lead to the formation of a more diverse chemical composition of subpermafrost water with a relatively low concentration of fluoride compared to that in aquifers on the left bank of the Lena River.

Minimal fluoride concentrations are measured in groundwater sampled in boreholes close to the main Lena channel regardless of the lithological composition of water-bearing rocks. Thus, fluoride concentrations in subpermafrost water average 2–3 mg/L in areas of the Tabaga and Kangalass promontories, do not exceed d 1.6 mg/L around the Sangary settlement and decline to 0.4–0.8 mg/L around the Kytyl-Dura and Oktemtsy settlements. Such a low fluoride concentration is due to the presence of open talik zones that probably cause the recharge of subpermafrost water through infiltration of river water. This assumption is confirmed by synchronicity of fluctuations in the water levels of the Lena River and subpermafrost water under the Lena River floodplain in the area of the Tabaga settlement [*Shepelev et al., 2002*].

CONCLUSIONS

Fluoride concentrations in subpermafrost aquifers of Central Yakutia almost everywhere exceed the norms for drinking water. Fluoride concentrations in subpermafrost water are largely controlled by hydrodynamic and geocryological processes near the bottom of the cryogenic aquiclude. A specific geochemical environment favoring fluorine extraction from water-bearing rocks and its accumulation in the solution is formed under their influence. The groundwater of bicarbonate–sodium composition with high alkalinity is maximally enriched with fluorine. Changes in the permafrost thickness under conditions of impeded water exchange are the reason for unstable equilibrium between groundwater and enclosing rocks, which ultimately contributes to the accumulation of reactive chemical elements, including fluorine, in the water.

Such geological conditions were formed within the Yakutsk uplift of the crystalline basement, where the Archean formations are overlain by the Lower and Middle Jurassic terrigenous aquifers. Here, there is a high probability to observe the considerable fluoride concentrations in subpermafrost water.

Optimal fluoride concentrations are only observed in areas, where aquifers are hydraulically connected with the Lena River through open taliks.

The identified distribution pattern of fluoride should be taken into account while setting up prospecting and evaluation of subpermafrost water to eliminate problems with the drinking water supply for the population and planning appropriate water treatment measures.

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