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SURFACE AND GROUND WATERS IN TERRESTRIAL PERMAFROST REGION

THE CHEMICAL COMPOSITION OF WATER OF LARGE EAST SIBERIAN RIVERS AND ITS DEPENDENCE ON THE THICKNESS OF SEASONALLY THAWED LAYER IN THE CATCHMENTS

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The main features of the chemical composition and physical characteristics of water in twelve largest rivers of East Siberia (Lena, Vilyui, Kolyma, Aldan, Olenek, Vitim, Indigirka, Amga, Olekma, Anabar, Yana, and Chara) were determined on the basis of observations during summer low-water runoff in 2007–2011. It was found that favorable oxygen regime, relatively high values of the chemical oxygen demand and water color, higher concentration of total iron and ammonium ions, and moderate salinity are characteristic of the studied rivers. East Siberia is a region with a ubiquitous distribution of permafrost. The thickness of seasonally thawed layer within river catchments is extremely variable. Using canonical-correlation analysis, it was found that concentrations of specific components of ionic constituents (water hardness, calcium, magnesium, bicarbonates, sulfate ions, and salinity) depend on the active layer thickness (ALT). Herewith, the deeper the active layer in a catchment, the higher the concentrations of these ionic constituents in river water. This is explained by the fact that permafrost serves as a barrier preventing infiltration of surface water into deep mineral horizons and thus limiting water saturation with mineral ions.

Keywords: *physico-chemical composition of water, major ions, salinity, permafrost, seasonally thawed layer, large rivers, East Siberia.*

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INTRODUCTION

East Siberia is characterized by a ubiquitous distribution of permafrost. Permafrost thickness is found to be 350–450 m in the central part of the region and reaches 1500 m further to the north, in the Olenek River basin [*Shepelev, 2009*]. The seasonally thawed layer (STL) varies in thickness from 0.1 to 1.5 m in coastal areas in the north of the region and increases to 3 m in the southern part, in the boreal zone [*Desyatkin et al., 2009*]. In the central part of the region, seasonal thawing begins at the end of April and reaches its maximum at the end of August. In October, freezing occurs simultaneously from the top and from the bottom; upper and lower freezing layers together at a depth of 0.7–0.8 m at the end of November or in December [*Matveev et al., 1989*].

The STL has a significant potential to influence the chemical composition of surface water in permafrost areas [*Smith et al., 2005; Frey, McClelland, 2009*]. The mechanisms of such influence may be quite different. First, soluble chemical components in the upper permafrost become labile, when they are found in the STL. Second, in catchments with a deep STL, the depth of surface runoff and the duration of water being in the soil differ from those in the areas with a thin STL. These factors cannot but influence the surface water chemistry through exchange reactions between water and soil. Finally, the chemical composition of river water is influenced by the balance between precipitation and groundwater in river alimentation, which depends on the STL thickness in river catchments.

In modern conditions of global climate change, the relevance of issues related to the study of the STL impact on the chemical composition of river water is increasing. According to the results of STL dynamics modeling [*Stendel, Christensen, 2002*], it is likely that STL thickness will increase by 30–40% in a larger part of permafrost areas in the northern hemisphere by 2100. For some regions, such as East Siberia, where permafrost is ubiquitous, transformations caused by permafrost degradation processes can be most dramatic due to their scale, since they will cover the entire territory. Therefore, understanding of the mechanisms of the STL influence on the chemical composition of river water is quite significant for predicting the chemical composition of Arctic rivers and the rate of runoff of dissolved substances into the Arctic Ocean.

The purpose of this study is to characterize the main features of the chemical composition and physical characteristics of the water of large rivers in East Siberia and to measure an impact of STL thickness in river catchments on the formation of the chemical composition of river water.

MATERIALS AND METHODS

This study focuses on catchments of twelve largest rivers in East Siberia: Lena, Vilyui, Kolyma, Aldan, Olenek, Vitim, Indigirka, Amga, Olekma, Anabar, Yana, and Chara (Fig. 1). Water samples were taken from the upper water layer (0–0.3 m) near the banks and along the fairway during summer low water (June–August) in 2007–2011. Conservation and storage of water samples were carried out in accordance with generally accepted methods [*Semenov, 1977*].

Water transparency was determined using a Secchi disk; color, by photometric method on an SF-26 device; pH value, by the potentiometric method using a Multitest IPL-101 device; suspended substances, by gravimetric method; oxygen concentration, by titrimetric method; oxygen saturation percentage, by calculation method; carbon dioxide concentration, by titrimetric method with phenolphthalein; water hardness and calcium concentration, by titrimetric method; magnesium concentration, by calculation method; sodium and potassium concentrations, by atomic emission spectrometry using an AAnalyst400 AAS device; bicarbonates, by back titration method; chloride ions, by the mercurimetric method; sulfate ions, by the turbidimetric method on an SF-26 device; mineralization (sum of dissolved salts), by calculation method; ammonium ions, by the photometric method with Nessler reagent on an SF-26 device; nitrite ions, by the photometric method with Griess reagent on an SF-26 device; nitrate ions, by the photometric method with salicylic acid on an SF-26 device; phosphate ions and silicon, by the photometric method with ammonium molybdate on an SF-26 device; total phosphorus, by the photometric method with ammonium persulfate on an SF-26 device; total iron, by the photometric method with sulfosalicylic acid on an SF-26 device; biological oxygen demand (BOD_5), by the titrimetric method (iodometric determination); chemical oxygen consumption (COD), by the photometric method using a Fluorate-02 device; petroleum products, phenols, and anionic surfactants (AS), by the fluorimetric method on a Fluorat-02 device. Classifications generally accepted in hydrochemistry were used to characterize river water [*Alekin, 1953, 1970*]. A system of maximum permissible concentrations for piscicultural water use MPCpw was applied to assess water quality.

Full data on the physical and chemical properties of water at each sampling point of the studied rivers were published previously [*Gabyshev, Gabysheva, 2018*].

Data on the STL thickness (minimum, average, and maximum) were extracted using ArcGIS software in accordance with the coordinates of observation points (Fig. 1) from a georeferenced dataset. This dataset is based on materials from 1960–1987 and was published as an appendix to the work of K. Beer with coauthors [*Beer et al., 2013*]; it is available in the PANGEA repository as a NetCDF file. The Russian map "*Seasonal Freezing and Thawing of Soils*" was also used [*National Atlas..., 2001*].

The data array generated by the authors includes two groups of quantitative variables: physical and chemical indicators of water (28 variables) and characteristics of the seasonally thawed layer (3 variables). The total number of observations in the array is 303.

The method of canonical correlations for a paired set of quantitative characteristics [*Afifi, Azen, 1982*] describing the STL and the physicochemical parameters of river water was applied to analyzing multiple correlations. The use of a multidimensional model for the analysis of canonical values also made it possible to distinguish data of the studied array according to a grouping characteristic of the studied rivers. When testing statistical hypotheses, the critical level of statistical significance was assumed to be 5%. Statistical analysis procedures were performed in the Statistica 10 software package.

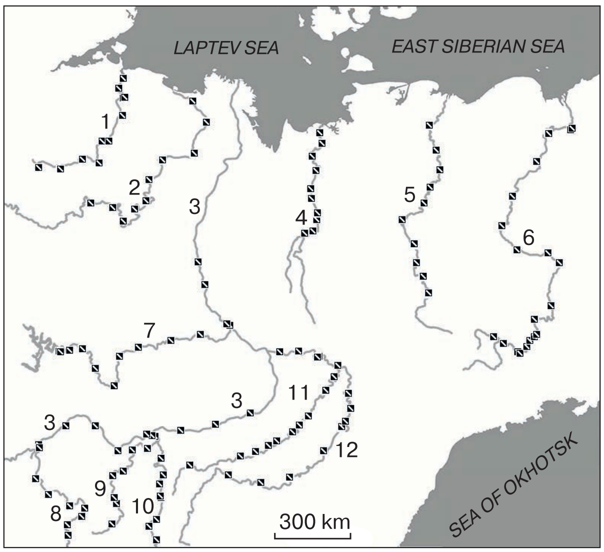

Fig. 1. Schematic map of the studied region and observation points (squares).

Rivers: 1 – Anabar, 2 – Olenek, 3 – Lena, 4 – Yana, 5 – Indigirka, 6 – Kolyma, 7 – Vilyui, 8 – Vitim, 9 – Chara, 10 – Olekma, 11 – Amga, 12 – Aldan.

RESULTS

The content of suspended substances in water from the Aldan, Olenek, and Kolyma rivers does not exceed 10 mg/dm^3 (hereinafter, average values for the entire river are provided). For the Yana, Anabar, and Indigirka rivers, it varies in the range from 25 to 65 mg/dm^3, for the Vilyui, Chara, Olekma, Lena, Vitim, and Amga rivers, it is 14–18 mg/dm^3. According to the hydrogen index, river waters are non-aggressive. The values of pH indicate a shift in the reaction to the neutral side (7.14–7.47) for the Aldan, Vilyui, Yana, and Kolyma rivers. The waters of the Chara, Olekma, Lena, Vitim, Amga, Anabar, Olenek, and Indigirka rivers are slightly alkaline (7.57–8.44).

Most rivers are characterized by an average oxygen content of 8.90–10.80 mg/dm^3 at 93–99% saturation. The waters of the Aldan, Yana, and Amga rivers are slightly oversaturated with oxygen; on average, the degree of saturation is 102–118% with an oxygen concentration of 9.90–11.50 mg/dm^3. In the Anabar River, the degree of saturation is characterized by lower values: 7.43 mg/dm^3 at 69% saturation, which also indicates a favorable oxygen regime. No cases of oxygen deficiency were recorded. The carbon dioxide content is low and varies within relatively narrow limits from 1 to 6 mg/dm^3.

According to the classification of O.A. Alekin, the waters of most of the studied rivers are fresh and low-mineralized (43.4 to 113.8 mg/L); in terms of hardness, they are very soft (from 0.49 to 1.28 meq/L). The exceptions are the Amga and Olenek rivers. Water of the Amga River is moderately mineralized (279.3 mg/L) and moderately hard (3.39 meq/L), and water of the Olenek River is also moderately mineralized (249.3 mg/L) and soft (2.84 meq/L).

All rivers are characterized by relatively low concentrations of major ions. The calcium content does not exceed 45 mg/dm^3, magnesium – 20 mg/dm^3, sodium – 10 mg/dm^3, potassium – 2 mg/dm^3, bicarbonates – 200 mg/dm^3, sulfates – 50 mg/dm^3, chlorides – 15 mg/dm^3. According to the classification of O.A. Alekin, waters of most of the rivers (Aldan, Vilyui, Lena, Vitim, Amga, Anabar, Olenek, and Kolyma) belong to the bicarbonate class, calcium group, type II–III. Waters of Chary, Olekma, Yana, and Indigirka rivers belong to the sulfate class, calcium group, type II–III.

The studied rivers are characterized by relatively high COD values and increased concentrations of ammonium and total iron. The average concentration of ammonium ions varies across rivers from 200 to 1095 μg/dm^3 (MPCpw 500 μg/dm^3); the color index varies from 7° to 57° (maximum permissible level 20°); total iron concentration varies from 0.04 to 0.98 mg/dm^3 (MPCpw 0.10 mg/dm^3); COD, from 11 to 58 mg/dm^3 (MPCpw 15 mg/dm^3).

The content of inorganic compounds is low: nitrites 11 μg/dm^3, phosphates 55 μg/dm^3, total phosphorus 150 μg/dm^3, nitrates 650 μg/dm^3, silicon 3 mg/dm^3. The concentration of phenols does not exceed 5 μg/dm^3; petroleum products, 45 μg/dm^3; surfactants, 157 μg/dm^3. The BOD_5 value is low – 1.50 mg/dm^3.

The studied rivers are characterized by a low excess of the MPC for COD (1.0–3.9 MPC), water color (1.0–2.9 MPC), and ammonium ions (1.0–2.2 MPC). An exception was noted for the total iron content. The detected total iron concentrations exceeded the MPCpw by 5–10 times in water of the Lena, Vitim, Indigirka, and Yana rivers and by 1.9–4.0 times in water of the Chara, Aldan, Vilyui, Olekma, Lena, Anabar, and Kolyma rivers.

The search for relationships between pairs of variables characterizing the physicochemical parameters of river water (28 variables, hereinafter referred to as the HYDROCHEMISTRY set) and the STL in the area of river basins (3 variables, hereinafter referred to as the STL set) was performed by the method of canonical correlations. As is known, the number of calculated canonical correlation coefficients corresponds to the minimum number of variables in one of the two analyzed sets. In our study, it equals to 3. For the most informative (first) solution, the canonical correlation R between STL characteristics and parameters of chemical composition of river water adjusted for the number of observations is 0.75. Thus, variables of the STL set and the HYDROCHEMISTRY set are strongly correlated. The significance level of the first canonical correlation has a value of $p < 0.0001$, therefore the obtained result is suitable for analysis. In applied research, four criteria (Table 1) are widely acceptable. They represent test statistics on the basis of which the researcher can make a conclusion on the null hypothesis. In addition, Table 1 shows the F-value (Fisher's test) and the Pr indicator (>F), which means the probability of the absence of correlation between the sets of variables. The Fisher test for each test statistic varies (Table 1), but Pr (>F) for each test is less than 0.05, so we reject the null hypothesis and conclude that the two sets of variables under study (STL and HYDROCHEMISTRY) are really correlated.

For a more detailed comparative analysis, normalized canonical coefficients, i.e., z-transformed variables with zero mean and one standard deviation were considered. The correlation coefficients between the variables of both sets (STL and HYDROCHEMISTRY) and the three canonical axes were calculated. Maximum correlations were found between the first canonical axes – most informative and interesting for further analysis. Table 2 shows the correlation coefficients between the STL variables and the canonical STL1 axis, ranked in descending order of their absolute values. These coefficients are standardized and dimensionless, so they are suitable for comparison with one another. The maximum weight in

Table 1. **Multivariate statistics of canonical correlation of variables in the STL (3 variables) and HYDROCHEMISTRY (28 variables) sets**

Statistic	Value	F value	Pr (>F)
Wilks' lambda	0.17	7.81	<0.0001
Pillai's trace	1.26	7.05	<0.0001
Lawley-Hotelling trace	2.65	8.56	<0.0001
Roy's largest root	1.55	15.20	<0.0001

Note: F is Fisher's test; Pr is probability value for Fisher's test.

Table 2. **Normalized canonical coefficients for the STL1 axis**

Variable: STL thickness, m	Weight of variable
Maximum	0.94
Minimum	0.15
Average	–0.04

the studied canonical axis is marked as the "maximum STL thickness". Other STL variables have insignificant weights.

Among the physicochemical characteristics of water, the most important ones are the hardness, total content of salts, and ionic composition (concentrations of calcium, bicarbonate, magnesium, and sulfate ions) (Table 3).

Thus, these components of the salt composition of water are closely related to the STL thickness. Moreover, the greater the depth of seasonal thawing, the higher the salt content in river water.

The data set was discriminated along the two obtained canonical axes for the rivers under study. In the scatterplot diagram (Fig. 2), the vertical axis collectively reflects a subset of hydrochemical variables, and the horizontal axis reflects the STL variables. Each point in Fig. 2 is a single observation (sampling point). The method of canonical correlations used in this study makes it possible to discriminate data between groups of observations. The authors discriminated data for the twelve rivers. In order to read the results of the analysis from a given scatterplot, it is necessary to evaluate the location of the observation points in the two canonical axes. The ordinate axis in the diagram corresponds to the canonical axis HYDROCHEMISTRY1, in which variables such as hardness and total salts have maximum weights. The X-axis corresponds to the canonical STL1 axis, where the variable "maximum STL thickness" shows the greatest weight. By assessing in which quarter of the diagram each point of observation is located, and where most of them are grouped, it is possible to determine the conditions that characterize the river in terms of maximum STL thickness, hardness, and degree of mineralization of water. Along the corresponding axis of the diagram, these parameters increase.

Table 3. **Normalized canonical coefficients for the HYDROCHEMISTRY1 axis**

Variable	Weight of variable
Hardness, meq/L	196.90
Ca^{2+}, mg/L	143.62
Total salts, mg/L	142.07
HCO^{3-}, mg/L	104.48
Mg^{2+}, mg/L	88.68
SO_4^{2-}, mg/L	27.94
Cl^-, mg/L	7.09
Na^+, mg/L	5.05
Phenols, μg/L	1.09
K^+, mg/L	–0.75
Si, mg/L	0.38
NH_4, mg/L	–0.36
CO_2, mg/L	0.29
AS, mg/L	0.22
COD, mg/L	0.22
BOD_5, mg/L	0.17
Oxygen saturation, %	–0.16
PO_4, μg/L	0.12
Water color, degrees	–0.12
Water transparency, m	–0.11
pH, units.	–0.10
NO_3, mg/L	–0.10
Fe_{tot}, mg/L	–0.10
O_2, mg/L	0.09
NO_2, μg/L	0.09
Suspended solids, mg/L	–0.04
P_{tot}, μg/L	–0.04
Oil products, mg/L	0.03

The results of observations along the studied rivers in these two axes clearly illustrate the fact that the Arctic rivers flowing into the Arctic Ocean such as the Olenek, Indigirka, and Yana rivers flow through territories with a low STL thickness (Fig. 2). The rivers in the central part of the region (Amga, Olekma, Aldan, and Vilyui) are located in the area with a thick STL. Rivers that cross significant distances in the meridional direction (Kolyma, Lena), as well as southern rivers (Vitim and Chara), the upper reaches of which are located in mountainous regions, in their different sections flow in the areas with different thickness of the STL. For the Anabar River, the sampling points were divided along the STL1 axis into two groups. Some of them characterize the areas with a thin STL, and some are shifted along the abscissa axis to the left of mark 0. This suggests that some of the sampling points on this river belong to the areas with a thicker STL. It is also evident that the rivers with the highest concentrations of salt components are located in the central part of the studied region (Amga, Olekma, Vilyui, and Aldan rivers).

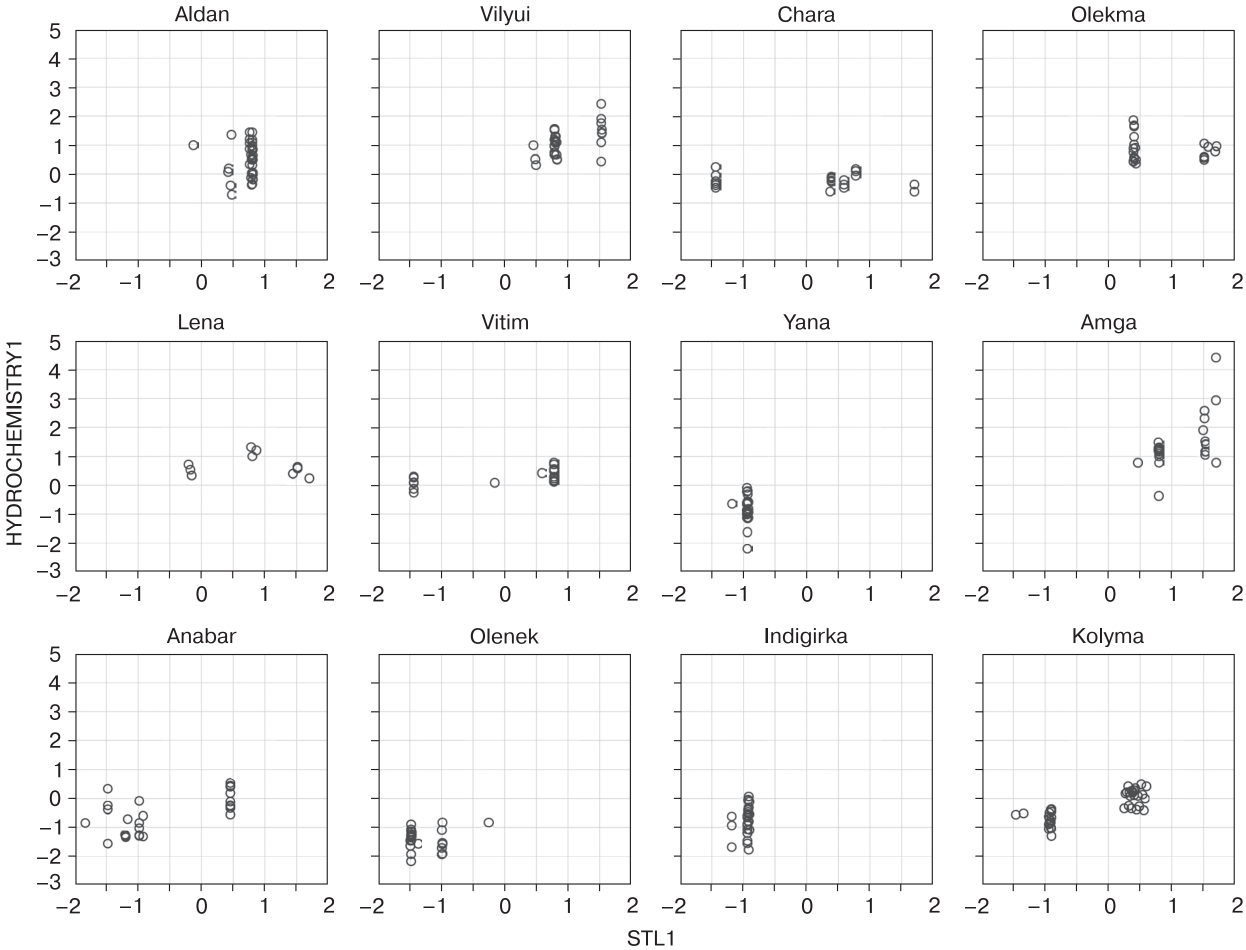

Fig. 2. Scatterplot of observations categorized by the studied rivers in the canonical axes HYDROCHEMISTRY1 (28 variables) and STL1 (3 variables).

DISCUSSION

For the studied large rivers of East Siberia, a favorable oxygen regime is typical. Most of the rivers are characterized by the low salt content. It is known that groundwater plays a significant role in the feeding of the Amga River compared to other rivers in the region [*Savvinov et al., 2000*]. This may explain the increased water hardness in this river. Relatively high COD and color values and increased concentrations of total iron and ammonium ions are characteristic of many reservoirs and watercourses in the region and are caused by natural factors [*Kirillov et al., 1979; Venglinsky et al., 1987*]. This phenomenon is associated with the entry of substances of natural origin into surface waters as a result of leaching from ferromanganese, copper pyrite, and other ores, as well as a result of the decomposition of bottom sediments [*Zenin, Belousova, 1988*].

The results of the canonical correlation indicate the relationship between the STL thickness and the concentration of a number of components of the salt composition of river water, namely hardness, sum of salts, calcium ions, bicarbonates, magnesium, and sulfate ions. The STL is characterized by low values in the north of the region and in the mountainous areas in the south. In the central part of the region, the STL is generally deeper [*Beer et al., 2013*]. A comparison between the patterns of spatial distribution of STL in the catchments of East Siberian rivers and the analyzed characteristics of the physical and chemical composition of river water made it possible to identify the following pattern: the deeper the base of STL of permafrost-affected soils in the drainage area, the higher the concentration of salt components.

The highest concentration of the six listed salt components was observed for four rivers in the central part of the region (Amga, Olekma, Vilyui, and Aldan rivers) flowing through the territory with the deepest STL. On the contrary, the majority of Arctic rivers confined to the catchments with thin STL are characterized by low concentrations of salt components.

This pattern can be explained using elements of the conceptual model of the STL influence on the chemical composition of surface water developed by R. MacLean [*MacLean et al., 1999*]. In spring, surface runoff is limited by the upper soil layers regardless of the presence of permafrost or seasonal frost. However, later, closer to the summer low-water period, such a factor as the STL thickness greatly influences the depth of penetration of runoff water flows into the soil. The conceptual model in question is based on the well-known fact that the upper soil horizon is characterized by accumulation of organic matter, and the underlying layer is mineral. A shallow STL keeps runoff flows close to the surface preventing them from penetration into the mineral soil horizons. A deep STL leads to a reduction in the time of contact of infiltrating surface water with the upper soil horizon and provides suprapermafrost runoff through underlying horizons rich in mineral substances [*Frey, McClelland, 2009*]. This difference in surface runoff pathways affects the chemical composition of water due to exchange reactions between soil and water [*Colombo et al., 2018*]. Thus, the STL thickness influences the natural process of transfer of soluble substances from soils to rivers with surface runoff.

A comparative study conducted on watercourses in the Western Siberia also showed that permafrost prevents the saturation of river water with mineral components. Thus, it was found that the total content of inorganic dissolved substances defined as the sum of eight components (Ca^{2+}, K^+, Mg^{2+}, Na^+, Si, Cl^-, HCO_3^-, SO_4^{2-}) averages 289 mg/L for rivers with catchments without permafrost and 48 mg/L for rivers located in the permafrost zone [*Frey et al., 2007*]. The authors associate this difference in the total mineralization of water with the hydrological features of the catchments, which depend on the presence of permafrost. According to researchers, permafrost forms a barrier that prevents the infiltration of surface runoff into deep mineral horizons, and also limits the influx of mineral-enriched sub-permafrost groundwater into surface water [*Woo, Winter, 1993; Michel, van Everdingen, 1994; Woo et al., 2000*].

This hypothesis is confirmed in other studies. Using the example of Alaskan watersheds with isolated permafrost, a higher content of bicarbonates, sulfates, as well as calcium, magnesium, potassium, and sodium ions was established in rivers flowing in areas of watersheds devoid of permafrost in comparison with adjacent areas of the permafrost zone [*Stottlemyer, 2001; Petrone et al., 2006, 2007; Keller et al., 2007*]. A significant decrease in the concentration of Ca^{2+} and Mg^{2+} ions was shown for taiga streams in Alaska influenced by permafrost distribution, which limits runoff within the upper soil horizon [*MacLean et al., 1999*].

Studies conducted in the watersheds of Central Siberia indicate that the concentration of calcium, sodium, magnesium, and potassium ions in river water increases significantly in the summer compared to the spring. Researchers attribute this to the fact that the mineral horizons become part of the STL in the summer, so that runoff flows penetrate into them, and these horizons become the source of mineral ions entering runoff flows and, finally, river water [*Parham et al., 2013*].

Comparative geochemical studies of the modern STL and the top of permafrost show that the upper permafrost layer is richer in minerals in relation to the overlying STL, which is due to a gradual removal of dissolved substances from the STL [*Kokelj, Burn, 2005*]. This indirectly confirms the validity of the above-mentioned conceptual model.

CONCLUSIONS

The catchments of East Siberia are characterized by the widespread distribution of continuous, discontinuous, and sporadic permafrost. The STL thickness in permafrost-affected soils greatly varies within the region. This is a factor that controls the natural transfer of soluble substances from soils to rivers with surface and soil runoff. The authors found that the studied rivers of the region are characterized by a favorable oxygen regime, relatively high COD and color values, increased concentrations of total iron and ammonium ions, and a low salt content.

The chemical composition of river water is formed in a complex natural system under operation of many factors, including the lithological composition of the soils in river catchments and the rocks of river beds, under-channel taliks with pressure waters, relief features controlling the speed of river flows and, hence, the time of interaction between river water and underlying sediments, etc.

In this study, we considered the influence of the STL thickness on the chemical composition of river water in permafrost landscapes of East Siberia. It was found that the STL thickness in the catchments is correlated with some hydrochemical characteristics of river water, such as hardness, the sum of salts, and concentrations of calcium, magnesium, bicarbonate, and sulfate ions. In the catchments with a thicker STL, the concentration of these salt components in river water is higher in comparison with rivers flowing in areas with a shallower STL. Rivers in the central part of the region (Amga, Olekma, Vilyui, and Aldan) flowing through the catchments with a thick STL are characterized by the highest concentrations of the salt components. The revealed pattern is consistent with the main provisions of the conceptual model of the STL influence on the chemical composition of river waters.

In recent decades, an increase in the STL thickness in permafrost-affected soils has been observed throughout the entire permafrost area in the northern hemisphere. Further increase in the STL thickness in the 21st century is predicted. In this regard, our results are important for assessing possible changes in the chemical composition of river water in the Arctic regions in the future and the entry rate of dissolved substances into the Arctic Ocean.

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