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Hydrochemical composition of thermokarst lake waters in the permafrost zone of western Siberia within the context of climate change

R. M. Manasypov[1,2], **O. S. Pokrovsky**[1,3], **S. N. Kirpotin**[2], **and L. S. Shirokova**[1,3]

[1]Geoscience and Environnement Toulouse, UMR 5563 CNRS, Université de Toulouse, 14 Avenue Edouard Belin, 31400, France
[2]Tomsk State University, 634050, Tomsk, 36 Lenin av., Russia
[3]Institute of Ecological Problems of the North UroRAS, 163061, Arkhangelsk, Nab. Severnoj Dviny, 23, Russia

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Correspondence to: O. S. Pokrovsky (oleg@get.obs-mip.fr)

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Abstract

This work describes the current state of thaw lake and pond ecosystems, the mechanisms of their formation and succession, which was assessed via field work during several summer campaigns, and the laboratory analysis of water samples that were collected in the northern part of western Siberia within continuous and discontinuous permafrost zones. We analyzed the elemental chemical composition of lake waters as a function of lake diameter, over more than two orders of magnitude of the lake size, and described the peculiarities of the elemental composition of the thermokarst water body ecosystem during various stages of lake development. We revealed significant correlations between Fe, Al and dissolved organic carbon (DOC) and various chemical elements across a latitude gradient of approximately 900 km. Several groups of chemical elements were distinguished that reflect the dynamic succession of the studied area of water bodies. Combining the data of the studied latitude profile with the information available in the current literature demonstrated that the average dissolved elemental concentrations in lakes of different size ranges exhibit specific dependencies on the latitude position, which is presumably linked to (1) leaching of the elements from frozen peat, which is the main source of solutes in thermokarst lakes, (2) marine atmospheric aerosol depositions, notably at the proximity to the sea border, and (3) short-range industrial pollution of certain metals from the largest Russian arctic smelter. We discuss the evolution of thermokarst lake chemical compositions during their formation and drainage and foresee the consequences of climate warming and permafrost thaw on the hydrochemistry of the thaw lakes and ponds of western Siberia.

1 Introduction

The wetland ecosystems of the subarctic region of western Siberia act as unique natural indicators of climate change because these ecosystems are most sensitive to alterations in the natural environment. The main reason for this sensitivity is the presence of

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frozen peat deposits, which become highly unstable under climate warming scenarios and thermokarst development (Kirpotin et al., 1995, 2003, 2008; Kirpotin and Vorobiov, 1999; Kravtsova and Bystrova, 2009; Callaghan et al., 1999; Muldiyarov et al., 2001). Until recently, the majority of studies on thermokarst, i.e., the thawing of ice-rich permafrost or the melting of massive ground ice (i.e., Jorgenson et al., 2008), have been performed in Canada, Alaska, Mongolia, China, Antarctica and eastern Siberia (see Kokelj and Jorgenson (2013) for a review and Boike et al. (2013) for a description of the Lena River Delta Observatory), with relatively little attention to the vast area of western Siberia where the largest resources of potentially highly unstable frozen peat resources are concentrated. The majority of frozen peat in this region is present as palsa-like raised bog complexes that are subjected to permafrost thaw and lake formation. Thermokarst lake formation is a dominant mode of permafrost degradation that consists of a surface disturbance, the subsequent melting of ground ice, surface subsidence and water impoundment (Grosse et al., 2013). The concept of the cyclic development of raised bog complexes was suggested by Scandinavian researchers (Matthews et al., 1997; Sollid and Sorbel, 1998), and further developed for western Siberia several years ago (Kirpotin et al., 2007, 2008, 2011). Briefly, the process of soil thaw starts from permafrost subsidence at the flat bog with the formation of a soil depression, which is an "embryo" of the thaw lake that continues growing and increasing its size until it achieves the km-size mature stage. At this stage, the lake may be drained via connecting to another water body or to a hydrological network, thus forming the drained lake or khasyreys. On the dry bottom of khasyreys, the frozen peat bogs starts to thaw, thus initiating the new cycle of thermokarst lakes. The described process may be well traced on satellite images; the analysis of this information allows the conclusion that, currently in the north of western Siberia, the dominant processes are permafrost degradation and the increasing number of small thaw lakes (Kirpotin et al., 2009; Bryskina and Kirpotin, 2012).

Thermokarst lakes of western Siberia are relatively poorly studied, with only a few studies reporting their hydrochemical composition (Savchenko, 1992; Leonova, 2004;

Leonova et al., 2005; Moskovchenko, 2010; Pokrovsky et al., 2011, 2013; Savichev et al., 2011; Manasypov et al., 2012; Shirokova et al., 2013). Within the northern border of the discontinuous permafrost, a limited number of thaw lakes and ponds near the town of Novyi Urengoy have been investigated (Shirokova et al., 2009; Audry et al., 2011). The major goal of the present study was to extend the coverage of permafrost lake sampling much further to the north, in the continuous permafrost and coastal regions, to reveal conceptually novel features of landscape and latitude control on the hydrochemistry of thaw water bodies. We anticipate that achieving this goal should allow us to quantify the average dissolved organic carbon (DOC), major and trace element (TE) concentrations and stocks in the thermokarst lakes all across the permafrost-bearing zone of western Siberia and to predict the magnitude of possible changes of these parameters using the "substitute space for time" approach (Frey et al., 2007). Frey et al. investigated a suite of large and small rivers and streams that were located from 55° N to 69° N of the West Siberian Plain and demonstrated a distinct change in the stream water hydrochemistry from the permafrost-free zone, where the rivers are fed by groundwater, to the permafrost-bearing zone, where the main feeding is due to surface peat dissolution. However, no significant gradient was found in the stream water chemical composition within the permafrost zone, northward of 61° N. The thermokarst lakes of this study, which are located exclusively within the permafrost-bearing zone, may offer a new dimension to the evaluation of the permafrost effect on surface water hydrochemistry. Indeed, the lakes better integrate multiple processes at the landscape level and have significantly higher spatial resolution due to their much smaller watershed areas than those of rivers and streams. For this purpose, the West Siberian Plain presents an ideal case study given the following characteristics: (1) contrasting permafrost coverage, from sporadic to continuous, (2) homogeneous geomorphological/orographic context; (3) high homogeneity of peat soil and underlain Neocene clays and sands over a significant latitude profile and (4) relatively easy access to study sites thanks to the existing infrastructure, which is linked to the oil and gas industry.

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Based on the recently collected and available literature data, we aimed to answer the following questions:

(1) Is there a latitude gradient of DOC and TE concentrations in thermokarst lakes that are at the same stage of development (subsidences, ponds, large lakes and drained lakes)?

(2) Is it possible to establish a functional dependence between the dissolved lake water component concentration and the lake surface area and to extend these dependences for the whole territory of the western Siberia permafrost zone?

2 Study sites and methods

The studied region is located within a tundra and forest-tundra zone of the northern part of western Siberia wetlands, within the discontinuous and continuous permafrost zones (Fig. 1). This region is dominated by the presence of small thermokarst (thaw) lakes of less than 1 km^2 surface area; their total surface area is estimated at 44 000 km^2 (Rikhter, 1963; Domaniczkii et al., 1971; Ivanov and Novikov, 1976; Kravtsova and Bystrova, 2009). The water objects investigated in this work are primarily thermokarst lakes that are located within peat sphagnum bogs; the bottom sediments of the lakes are dominated by peat detritus. Active thermokarst occurs due to the thawing of syngenetic and epigenetic segregation ice, ice wedges and ice layers in the deep (> 2 m) horizons and primarily due to the ice thawing of the active layer (< 2 m). Because of thermokarst activity, various negative relief forms are developing: depression, subsidences and ponds, which are usually separated by flat mound peat bogs up to 2 m in height (Kozlov, 2005). The largest thermokarst lakes that are located within the peat bog are km-size, with a depth of 0.5–1.5 m (Savchenko, 1992; Kozlov, 2005).

All sampled thermokarst lakes of western Siberia were separated into five stages of evolution, depending on the lake diameter and, presumably, on the age, as follows: 2 to 10 m, 11 to 100 m, 101 to 500 m, > 500 m and drained lakes (khasyreys) to 5000 m, which correspond to 1st, 2nd, 3rd, 4th and 5th stages, respectively. The separation of

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these different stages was based on the already described chronosequence of lake formation and cyclic development (cf., Kirpotin et al., 2007, 2008; Pokrovsky et al., 2011). The lake diameter was determined on-site using a GPS navigator and verified from the topographical and Google Earth® maps. The sampling was performed within three major study sites, as depicted in Fig. 1: (1) the continental zone, including the district of Pangody (Yamalo–Nenetsk Region), west of the Novyi Urengoy region in August 2010; (2) east of the Novyi Urengoy region in August 2011, and (3) the coastal zone, around the village of Gyda in August 2011. Sites 1 and 2 lay within the discontinuous permafrost zone and site 3 is located within continuous permafrost zone. Using all-terrain 8-wheels vehicle (ARGO 8x8 700 HD), we sampled in total 43 thermokarst lakes of the continental zone and 15 lakes of the coastal artic zone. To characterize the variability of thermokarst lakes along a ∼ 1000 km latitude profile, previously collected data on lake water hydrochemistry in the southern part of the cryozone (Nojabrsk region, Shirokova et al., 2013) were also used. Note that because of hydrocarbon extraction, the surface water and groundwater may be contaminated with oilfield brines (e.g., Collins, 1975; Alexeev et al., 2004). However, all the studied lakes and thaw ponds lay outside the oil extraction fields. Thaw lakes are located within the largest natural gas provinces which present relatively little impact on surface ecosystems.

Water samples were collected from the surface (0.3 to 0.5 m) of shallow (0.5 to 1.5 m depth) lakes in pre-cleaned polypropylene 250 mL containers and filtered on-site or within 4 h after sampling through disposable MILLEX Filter units (0.45 µm pore size, 33 mm diameter) using sterile plastic syringes and vinyl gloves. The first 20–50 mL of the filtrate was discarded to allow for filter conditionings. The filtered sample was divided into two parts: one is acidified with bidistilled nitric acid for cation and trace element analyses and the other part is kept without acidification for DOC and anion analyses. The samples were kept in the refrigerator before the analyses; the effect of sample storage and conditioning on measured elements and DOC concentrations was found to be absent. Field blank experiments were routinely performed via filtering the MilliQ water and analyzing the final element concentration; these blanks were found to be < 5 % of typical concentrations for DOC and major cations and anions, < 10 % for divalent metals (Zn, Cu, Fe, Cd, Pb) and < 2 % for the other trace elements (TEs).

Concentrations of DOC, Cl^-, SO_4^{2-}, Alk, cations, and TEs were measured using methods that are routinely practiced in our laboratory for the analysis of boreal, organic-rich, low-mineralized water samples (see Vasyukova et al., 2010; Pokrovsky et al., 2010, 2011, 2012). TEs were measured using inductively coupled plasma-mass spectrometry (ICP-MS, Agilent 7500 ce) with indium and rhenium as internal standards and a precision of better than ±5 %. The international geostandards SLRS-4 and SLRS-5 (Riverine Water Reference Material for Trace Metals certified by the National Research Council of Canada) were used to check the validity and reproducibility of each analysis. A good agreement was obtained between our replicated measurements of SLRS-4 and the certified values, with a relative difference of < 10 % except B and P (≤ 30 %). In addition to these analyses, all lake water samples were measured using a high-resolution ICP MS (Element XR) with a factor of 100–1000 higher detection limit for most elements and a relative uncertainty of analysis between 5 and 10 %. This high resolution greatly increased the detection limit of many elements and improved the precision of the analyses while avoiding interferences. The average agreement between two ICP MS instruments for the majority of the TEs was within 10–15 %. The non-acidified sample was used for (1) Si determination with ammonium molybdate on an AutoAnalyzer 3 Bran + Luebbe with ±2 % uncertainty and a 10 $\mu g\,L^{-1}$ detection limit; (2) DOC analyses with total combustion at 800 °C using a Pt catalyzer (TOC-VCSN, SHIMADZU) with 5 % uncertainty and a detection limit of 0.1 $mg\,L^{-1}$; UV absorbency at 280 nm (spectrophotometer Varicen, Cary 50 Scan. UV-Visible); measuring chloride and sulfate using liquid chromatography (DIONEX ICS-2000) with an uncertainty of 2 % and a detection limit of 0.02 $mg\,L^{-1}$. Element concentrations and speciation data were analyzed with best fit functions based on the method of least squares, Pearson correlation and one-way ANOVA with the STATISTICA version 8 software (StatSoft Inc., Tulsa, OK). Regressions and power functions were used to examine relations between element concentrations and lake surface areas. Correlation coefficients were calculated

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to elucidate relations between organic carbon/Fe and TE concentrations in lakes. A criterion of a significant correlation between elements was Pearson coefficients that were higher than 0.5 ($R^2 \geq 0.5$). The ANOVA method was used to test the differences in average TE concentrations and DOC parameters as well as concentration – lake surface regression slopes for different geographic territories. The ANOVA test was carried out with a one-way analysis of variance by using the Dunn's method due to the different number of samples for each zone (SigmaPlot version 11.0/Systat Software, Inc). In this method, $P < 0.05$ indicates that the difference in the median values is important and statistically significant. In contrast, $P > 0.05$ indicates that the differences in the median values are not statistically significant and that these differences may stem from random sampling variability.

3 Results and Discussion

3.1 Major anions, conductivity and pH

All studied thermokarst subarctic lakes from the continental zone are primarily fed by the atmospheric source as determined from the analysis of major anions (Table 1). Chloride and sulfate concentrations range from 0.014 to 1.12 and from 0.04 to 1.1 $\text{mg}\,\text{L}^{-1}$, respectively, which matches the range of the values that are typical for atmospheric precipitates in this region (Granat, 1972; Savichev, 2005; Moskovchenko and Babushkin, 2012). Among all studied lakes, however, there is one lake that exhibits an elevated sulfate concentration (3.5 $\text{mg}\,\text{L}^{-1}$), and this lake has a different, deep blue color on the remote satellite images of the territory (not shown) (sample RM20, Table 1). This lake is most likely the primary deep lake, which is subjected to the influence of underground waters. Similar "blue" lakes were reported on the Tazovsky Peninsula (Kuzin et al., 2012). The total dissolved solid concentration of thermokarst lakes of subarctic (continental) western Siberia vary slightly among different sizes and stages of development, with an average of $2.7 \pm 1.1\,\text{mg}\,\text{L}^{-1}$.

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It is known that the surface waters of the southern part of Gyda Bay are enriched in Na^+ and Cl^- (Tomberg et al., 2011). Indeed, the lakes of the coastal arctic zone that were sampled in this work contain from 0.6 to 6.9 $mg\,L^{-1}$ of chloride and from 0.02 to 1.6 $mg\,L^{-1}$ of sulfate. The large variability of these ion concentrations likely reflects the different degree of marine aerosol influence, which is illustrated in Fig. 2, where chloride concentration is presented as a function of lake diameter for discontinuous and continuous permafrost zones. The total dissolved salt concentration of thermokarst lakes of the coastal arctic zone of Western Siberia is equal to $9.0 \pm 5.2\,mg\,L^{-1}$ and ranges from 3.3 to 22.7 $mg\,L^{-1}$.

The analysis of the conductivity of thermokarst lake waters from the continental zone, which reflects total dissolved solid content, does not demonstrate any significant variation among the different stages of lake development, although the maximal values of conductivity ($\sim$ 20 to 30 $\mu S\,cm^{-1}$) are recorded on the 1st and 2nd stages. Overall, the studied waters have low mineral content, with the typical conductivity ranging from 10 to 17 $\mu S\,cm^{-1}$.

In the Arctic coastal zone, the conductivity is significantly higher (73 $\mu S\,cm^{-1}$ on average, with a range from 40 to 250 $\mu S\,cm^{-1}$), which corresponds to higher major ion concentrations (Fig. 3). Disregarding a small depression, which presumably has marine aerosol influences, the coastal lakes of Gyda exhibit an increase in conductivity with an increase in the lake size (Fig. 3). This observation may be linked to increasing connections of large lakes to the groundwater network and to saline deep waters, given the proximity to the sea. These groundwaters are absent in the southern part of studied territory, which is far from the coast. From Fig. 3, the studied thermokarst lakes exhibit the lowest specific conductivity in comparison with other permafrost thaw water bodies that are reported in the literature, such as the Lena Delta (Abnizova et al., 2012). Such a low specific conductivity of western Siberia thermokarst lakes of the continental zone stem from the organic, and not the mineral, nature of the surrounding substrate.

The pH of thermokarst lakes from the continental site ranged from 3.9 to 5.4; generally, the 1st stage exhibited more acidic conditions compared with the other stages

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of lake development (Fig. 4). This higher acidity stems from the impact of acidic peat leaching on the lake water chemistry, which is more pronounced at the beginning of thawing, at high solid substrate/surface fluid ratios. This observation is illustrated by a distinct trend of increasing pH with increasing lake size in this region. In contrast, the coastal lakes of the arctic zone, which are located within clay and sand bedrocks, are influenced by spring flooding and by the proximity to the sea and thus exhibit a significantly higher pH that ranges from 6.2 to 6.6. Therefore, the majority of the studied western Siberian thermokarst ponds and lakes are significantly more acidic than the other thermokarst water bodies in North America, Fennoscandia or Eastern Siberia. Only the coastal Gyda site exhibits circumneutral pH values, which are certainly due to the effect of marine deposits as solid substrates.

3.2 Dissolved organic carbon

A high concentration of dissolved organic matter (DOM) is among the major specific parameters of studied thermokarst lakes. In the Russian boreal zone, organic matter leaching from peat and bog soils provides a significant concentration of DOM and a low concentration of inorganic ions (Alekin, 1953). As a result, the DOM often constitutes more than 50 % of total solutes, particularly in the northern part of western Siberia, such as the Tazovsky district (Savchenko, 1992). Overall, the DOC concentration of the studied lakes shown in Fig. 5 is within the trends that have been reported among the discontinuous permafrost zone of the Novyi Urengoy region (Pokrovsky et al., 2011), the discontinuous/sporadic permafrost region (Shirokova et al., 2013) and the upland lakes of the Mackenzie Delta region (Kokelj et al., 2005). At the same time, western Siberia thaw lakes are richer in DOC compared with lakes of the Canadian subarctic (Laurion et al., 2010; Bouchard et al., 2013) and the Lena Delta (Abnizova et al., 2012).

Analyses of thermokarst lakes from the continental zone demonstrated a higher concentration of DOC at the 1st and 2nd stages of development (18.1 ± 15.4 and 15.3 ± 8.1 $\text{mg}\,\text{L}^{-1}$, respectively) compared with the mature stages (11.0 ± 4.3 and 11.1± 3.4 $\text{mg}\,\text{L}^{-1}$, for 3th and 4th stages, respectively) (Fig. 5). Such a progressive de-

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crease in the DOC concentration during ecosystem development, which corresponds to thermokarst lake growth and maturation, may stem from the combination of two parallel processes: (1) the enrichment of the water column during peat soil leaching at the lake border and the thawing of soil surface ice, and (2) the heterotrophic mineralization of DOM by aerobic bacterioplankton in the water column. Both processes have been demonstrated to operate in thermokarst lakes of discontinuous (Pokrovsky et al., 2011) and sporadic/discontinuous (Shirokova et al., 2013) permafrost zones. Upon a progressive increase in the ratio of [water volume] to [board perimeter], the input of allochthonous DOM to the lake decreases, whereas the intensity of heterotrophic respiration of DOM into CO_2 remains constant, thus leading to a decrease in the DOC concentration during thaw lake ecosystem maturation. An additional reason for the decrease in the DOC concentration in large mature lakes may be the photo-oxidation of high molecular weight organic substances in the water column, which is fairly well-known in other boreal (Jonsson et al., 2001, 2008) and subarctic aquatic systems (Pokrovsky et al., 2011; Shirokova et al., 2013). After large lake drainage and khasyreys formation on the last, or 5th, stage of development, the DOC content increases again to $15.6 \pm 3.6\,\mathrm{mg\,L^{-1}}$. This increase may be linked to the presence of aquatic plants, phytoplankton and periphyton, which contributes to the production of autochthonous exometabolites, thus increasing the DOC level in the thermokarst lake water (Audry et al., 2011). Similar phenomenon also observed in the Mackenzie Delta lakes (Tank et al., 2009, 2011).

The DOC in the water column of the arctic lakes from the coastal zone of the Gyda region ranged from 3.2 to $21.3\,\mathrm{mg\,L^{-1}}$, with the highest concentrations that were encountered in the smallest (< 100 m diameter) water objects. The average DOC concentration was equal to $7\,\mathrm{mg\,L^{-1}}$, which is a factor of 2 lower compared with continental lakes. This difference is in general agreement with the trend of increasing DOC concentrations from the forest toward the forest-tundra and tundra zones, with minimal DOC concentrations observed in lakes that are located several hundred km north of the tree line (Vincent and Pienitz, 1996).

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3.3 Major and trace element concentration in thermokarst lakes

The specificity of studied water objects is that Fe and Al are among the major dissolved components in the water column and that the speciation of the majority of trace elements is controlled by their association with Fe, Al-rich organo-mineral colloids and allochtnous organic matter that originated from peat leaching at the border (Pokrovsky et al., 2011). The progressive change of the lake water chemical composition during lake cyclic development and maturation seems to be controlled by DOM mineralization via heterotrophic aerobic bacterioplankton, which leads to colloid coagulation and the transit of dissolved mineral components to the sediments (Audry et al., 2011). This observation suggests the existence of correlations between different elements that are involved in the same processes or, in the case of major and trace components, the capability of controlling each other. Examples of correlations between lake water major parameters (Al, Fe, DOC and specific conductivity) are shown in Fig. 6. Whereas Fe and Al exhibit a significant correlation with DOC over the full size of lakes and ponds both in continuous coastal and discontinuous continental sites, this correlation is not the case for specific conductivity, which is also correlated with DOC but exhibits two distinct pattern of dependencies, with the arctic coastal zone of continuous permafrost abundance being distinctly enriched in dissolved salts at a rather low range of DOC.

The highest concentrations of Fe, Si and DOC are observed at the beginning of lake formation, and the lowest concentrations of these components are encountered in large mature lakes, as illustrated on plots of major and TE concentrations as a function of the lake diameter (Fig. 7a–f). The arctic coastal lakes of continuous permafrost zone exhibit significantly higher concentrations of alkali and alkaline-earth elements compared with the continental (southern) thermokarst lakes (Fig. 7a–d). This observation is most likely due to the influence of sea salts, whether as marine aerosols or as underground feeding by saline waters. Notably, hosted rocks of the arctic lakes represented by Neocene marine clay deposits also contain a significant amount of easily exchangeable alkali and alkaline earth metals.

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Average measured element concentrations in the water column of studied thermokarst water objects from the southern (continental) zone are listed in Table 2 and can be classified as following, for the first ten most abundant elements:

1st, the initial stage of the thermokarst depression and soil subsidence (the diameter of the water surface is from 2 to 10 m): Fe > Si > Ca > Na > Mg > Al > K > Mn > Zn > Cr;

2nd stage (the diameter is from 10 to 100 m): Ca > Fe > Si > Mg > Na > Al > K > Mn > Zn > Cr;

3rd stage (the diameter is from 100 to 500 m): Ca > Na > Si > Mg > Fe > K > Al > Mn > Zn > Cr;

4th stage (the diameter is > 500 m): Ca > Na > Mg > Si > K > Fe > Al > Zn > Mn > Sr, and finally,

5th stage of drained lake or khasyreys: Ca > Si > Fe > Mg > Na > Al > K > Mn > Zn > Sr.

Therefore, one can observe a systematic evolution of the thermokarst lake chemical composition, from Fe-Si waters at the beginning of thaw lake formation to Ca-Na-Mg dominance in mature lakes of the 4th stage. The terminal 5th stage of lake development is characterized by the increase in the relative contributions of Si, K and Fe, which is likely due to the production and decay of macrophytes and phytoplankton in the remaining khasyrey's water bodies.

The arctic coastal lakes exhibit the following order of dissolved element concentration decrease: Na > Mg > Ca > K > Si > Fe > Al > Zn > Sr > Mn. Therefore, the studied lakes of the coastal continuous permafrost zone (Gyda) are similar to the mature thermokarst lakes of the 4th stage from the discontinuous permafrost, with a somewhat smaller contribution of Fe and Si and an increased contribution of Na due to the proximity to the sea.

The obtained data on the thermokarst lake composition allowed us to distinguish four groups of chemical elements, which reflects the dynamic succession of lake development, from the 1st to the last stage: (1) Element concentration decreases from 20 to

90 % during lake maturation: Cr, Cs, Mo, Ga; (2) Element concentration increases by 35 to 55 %: Ca, Li, Mg, Se; (3) Element concentration is the highest at the initial and terminal stages and is the lowest in mature thermokarst lakes of > 100 m diameter with the relative variation among different stages from 26 to 83 %: As, Co, Fe, Al, Si, Cd; (4) Element concentration is the lowest at the beginning and at the end of ecosystem development and is the highest in mature lakes that are larger than 100 m in diameter: V, U, Rb, B, K and major anions (Cl^-, SO_4^{2-}) with the relative variations between 32 and 83 %. The decrease in 1st and 3rd element group concentrations over the course of development of the lake ecosystem may be linked to the essentially colloidal status of these elements, which are present as organo-ferric HMW (1 kDa–0.45 µm) entities as determined by the size fractionation procedure that was applied to thermokarst lake waters of western Siberia (Pokrovsky et al., 2011; Shirokova et al., 2013). The decrease in DOC and Fe concentrations over the course of lake ecosystem development leads to a decrease in the concentration of allochthonous organo-mineral colloids, which bind these trace elements.

The other elements in the thermokarst lakes did not demonstrate any systematic variations as a function of lake stage development, and statistically significant differences among different stages are not demonstrated.

3.4 Trace element concentration dependence on lake diameter

The correlation coefficient between element concentration and lake diameter is element-specific and generally negative, i.e., the concentration decreases with the increase in lake diameter, as seen for DOC, Al, Fe, Mn, Co, Zr, REEs, Th (Table 3). Examples of typical relations between the element concentration and water body diameter are illustrated in Fig. 8a–d for several groups of trace elements: divalent metals (Co, Zn, Cd, Cu, Pb and Ni), oxyanions and neutral molecules (Mo, Sb, Cr and As), trivalent and tetravalent hydrolysates (Al, Ga, Ti, Zr) and soluble, marine aerosols/saline deposits – originated elements (B, Sr, Rb and U), respectively. For thermokarst lakes of the continental subarctic zone, clear tendencies of TE concentrations with the change in the water body size and significant differences in element concentrations at different stages of lake development with a similar size of the water body (i.e., 2nd stage and khasyreys) are observed.

Results from the arctic coastal lake zone follow the general tendency that was drawn by the data that were collected in the discontinuous permafrost zone of the continental region; however, there are also some peculiarities. For example, the concentration of the antimony is several times smaller in the arctic coastal lakes of the continuous permafrost zone compared with sites I and II of the discontinuous permafrost (Fig. 8b). In accord with significantly less acidic pH of the latter, the concentration of typical insoluble elements, such as Al or Zr, in > 100 m diameter lakes is significantly smaller in the Gyda region compared with the continental zone (Fig. 8c). Tetravalent hydrolysates (Ti, Zr, Hf, Th) do not exhibit any significant difference in their concentrations at different stages of lake development, beginning with the 2nd–3rd stage until the drained lake stage (khasyreys). This observation may suggest the absence of the visible external input of silicate material from underground sources or atmospheric dust deposits (Fig. 8c).

The decrease in element concentrations with the water body size increase are typical for the elements that are supplied to the lake water from the peat and plant litter leaching and that are then actively removed from the water column during chemical and microbiological processes without significant atmospheric or underground input. For example, this observation may be true for all divalent transition metals (Fig. 8a). Elements exhibiting the influence of atmospheric or underground input from sea salts are Rb, Sr, B and U (Fig. 8d). These elements demonstrate a concentration increase with lake diameter increase to the 4th stage, before lake draining.

To reveal the specificity of the chemical composition of thermokarst lakes at the beginning of their formation, we calculated the ratio of average element concentrations in the lakes of the 1st stage of formation (permafrost subsidences and depressions < 10 m diameter) to the average value of all other stages, which is illustrated as a stack diagram in Fig. 9. From this figure, we see that, at the beginning of lake formation, significant enrichment (≥ factor of 2) of the lake water by Fe, Si, Ga, Pb, Cd and insoluble trivalent and tetravalent elements that are bound to DOC and Fe occurs. Upon the development of the lakes and the increase of their size, Zn, Ba, Y, Ce, and La decrease their concentrations, whereas K, Rb, Se, Li, Cu, and, to a lesser degree, Mg, B, and Sb increase their concentrations. The significant increase in K (and its isomorphic Rb) upon lake maturation may be linked to the release of these elements from decaying plant litter and from aquatic macrophytes that are present at the final stage of lake evolution. Minimal changes in concentration were recorded for Na, V, Sb, U, Ca and Mn. These elements are not strongly associated with organo-ferric colloids. As such, these elements are not correlated with DOC, Fe or Al, which are the major colloidal components of the lake water that are subjected to significant transformation during the course of lake maturation. In addition, these elements do not have significant external sources (atmospheric aerosols, industrial activity) and, thus, act as highly conservative constituents of thermokarst lake water in western Siberia.

3.5 Correlations between elements in the thaw lakes water bodies

In agreement with previous investigations in the northern (Novyi Urengoy region, Pokrovsky et al., 2011) and southern (Nojabrsk region, Shirokova et al., 2013) discontinuous permafrost zones, there was a significant correlation between Fe, Al and DOC concentrations in the lake water. There was a lack of correlation between any of these elements and alkali (Li, Na, K, Rb, Cs), alkaline–earth elements (Mg, Ca, Sr, Ba) and anions (Cl^-, SO_4^{2-}), as well as trace elements that are not linked to organic or organo-mineral, such as Mo, Sb, B, Y, Mn, Ni, and W. Additionally, Si and As, although present in the form of neutral molecules, exhibited significant correlation with Fe rather than DOC ($R^2 = 0.84$ and 0.71, respectively, Fig. 10a and b). For Si, which does not have any colloidal form, this link may suggest the input of Si from the soil at the beginning of water body development, concomitant with that of Fe and its removal from the water column by diatoms or aquatic macrophytes at the terminal stage of the lake development cycle. The control of Fe colloids over As speciation in boreal and thermokarst lake waters is fairly well-known (Pokrovsky et al., 2012, 2013).

Among the divalent heavy metals, only Co, Cd and Pb exhibit a significant correlation with Fe or DOC (Fig. 10c–e). Whereas the concentration decrease for Co upon lake ecosystem maturation may reflect its uptake by growing phytoplankton cells and by scavenging to the sediments upon their death, the decrease of Pb concentration is likely to be linked to the decrease of its input from coastal peat abrasion during lake broadening, thus leading to a decrease in the ratio of lake border (representing the source provenance) to lake volume or lake surface (representing metal sink in the sediment). The significant accumulation of Pb in peat deposits is fairly well-known (Shotyk et al., 2000). Notably, the direct atmospheric (aerosol) deposition on the thermokarst lake water surface within the continental discontinuous permafrost sites should be rather small. If such an input was significant, then the concentration of these elements should remain rather constant for all stages (surface areas) of sampled water objects that all have similar depths (0.5 to 1.5 m). In addition, Cd exhibits two clusters of data points in its correlation with DOC (Fig. 10e). Almost a three-fold decrease in Cd concentration was observed both for arctic coastal and continental subarctic lakes during 3rd and 4th stages and in khasyreys. This likely reflects the change of metal source in the lake water depending on stage and the active biological uptake of this metal upon the lake maturation and lake size increase.

The rare earth elements (REEs) exhibited significant differences in concentrations in the thermokarst lake waters of the continental subarctic and arctic coastal zones. In the former, REEs correlate only with Al ($R^2 = 0.51$ and 0.59 for La and Yb, respectively), whereas in the latter, REEs correlate with DOC, Al and primarily with Fe ($R^2 = 0.50$, 0.88, 0.79 for La; $R^2 = 0.68$, 0.92, and 0.97 for Yb, respectively). Concentrations of REEs decrease with the increase in the lake size, similar to other trace metals that are controlled by organo-mineral colloids, as described above.

Overall, the observed correlation dependencies may reflect (1) the common source of materials, such as soil peat or deep silicate rocks, for Ca, Si, Al, Ti, Zr, Hf, Pb, and U

(Pokrovsky et al., 2011); (2) element-specific mobilization mechanisms in the form of organic and organo-mineral colloids, which are particularly visible for Fe, Al, trivalent and tetravalent hydrolysates (Shirokova et al., 2013); (3) processes of biological uptake and release (Zn, Mn, Co, Mo, Cu, Si, P, Cd) in the water column, which are particularly pronounced during hot summer periods (Pokrovsky et al., 2013); and (4) the sedimentation of solid phases containing TEs in the sediments and the diffusional flux to the bottom layers (As, Sb, Se, Cd, Pb) (Audry et al., 2011).

3.6 Comparison of TE concentrations in the thermokarst lakes with average world concentrations in surface waters

To the best of our knowledge, there is no comprehensive database on trace element concentrations in lakes; therefore, no “world average” value can be recommended. As such, we will use the available data for other boreal, subarctic and arctic lakes (Pokrovsky et al., 2011, 2012) and the average clark concentration in the river water (Gaillardet et al., 2003). The surface waters of the northern part of western Siberia present a low concentration of total dissolved solid and labile major and trace elements, such as Na, Mg, Si, K, Ca, Li, B, Mn, Rb, Sr, Mo, Ba, and U, which primarily originated from deep and shallow groundwater discharge during water-host rock interactions. Essentially, the peat (organic) nature of the hosting solid substrate for thermokarst lakes brings about elevated DOC and acidic pH waters compared with other thaw lakes that are located within the mineral substrates in the continental part of eastern Siberia (loesses) and the Alaskan arctic, particularly the Mackenzie Delta (i.e., Kokelj et al., 2009). TE concentrations in western Siberian thermokarst lakes are much higher compared with the following: (1) Al, Mn and Fe concentrations in the water column of the Imandra Lake of the Kola Peninsula (8, 10–22, and 17 $\mu g\,L^{-1}$, respectively, Ingri et al., 2011); (2) metal concentrations in the lakes of Canada’s Northwestern Territories (Pienitz et al., 1997a, b; Rühland and Smol, 1998; Lim et al., 2005; Côté et al., 2010), the Canadian High Arctic (Lim et al., 2001; Michelutti et al., 2002a,b; Antoniades et al., 2003) and arctic Alaska (Kling et al., 1992); and (3) the chemical composition of lakes in central Yakutia (Kumke et al., 2007; Wetterich et al., 2008) and northern Eurasia (Duff et al., 1999; Solovieva et al., 2005). There is significant difference in trace element composition between western Siberia thermokarst lakes and boreal, organic-rich lakes of non-permafrost zone (Pokrovsky et al., 2012). The majority of major and trace element dissolved in waters of thermokarst lakes from continuous and discontinuous permafrost zone are in lower abundances compared to typical boreal humic lake of the same level of DOC. Note that this conclusion is based on rigorous statistics of 50 water objects studied in this work and more than 7 yr of systematic measurements in the boreal non permafrost zone.

Elevated DOC in the form of allochthonous organic and organo-mineral colloids enhances the mobility of usually inert elements, such as trivalent and tetravalent hydrolysates; as a result, the transport of these elements from the soil substrate to the lake water increases. Compared with the world average concentrations in the river water, all stages of thermokarst lakes exhibit significant enrichment by elements whose migration in natural waters is controlled by organic and organo-mineral colloids, such as Al, Fe, Ti, Cr, Co, Ni, Zn, Y, Zr, Cs, La, Hf, and Pb (Fig. 11). This figure indicates that several metals, such as Ti, Cr, Ni, Cu, Zn, and La, are strongly accumulated in thermokarst lake waters compared with river waters.

3.7 DOC and metal concentration in thermokarst lakes of western Siberia across a 900 km latitude and permafrost profile

A straightforward way of predicting surface water changes under a climate warming scenario is by “substituting space for time”, which has previously been used for western Siberian rivers (Frey et al., 2007). Such an approach assumes that the contemporary difference on local and global scales within the climate gradient, which is created by latitude or by permafrost coverage and thickness, can serve as a proxy for future changes of a given system. The permafrost degradation leading to a shift from continuous to discontinuous and sporadic distribution will be the most important factor controlling the concentration of dissolved organic carbon and trace elements in western Siberia

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thermokarst lakes over next several decades. Indeed, given the large geographic coverage of the studied thermokarst lakes, i.e., ∼ 10° latitude along a strongly pronounced gradient of permafrost thickness from discontinuous and sporadic in the south of Nojabrsk district to continuous in the Gyda region, one can test the effect of climate or latitude position on lake water hydrochemistry. Note that such a representation may be partially biased by the fact that Gyda lakes are located within the proximity to the sea and, therefore, are subjected to the influence of arctic aerosol depositions. For this analysis, we used geometric mean values of element concentrations in lakes that were sampled during the summer period 2008–2012 for four size ranges of thermokarst water bodies: depressions smaller than 10 m diameter, thaw ponds less than 100 m diameter, small and medium lakes of 100 to 500 m diameter (the dominant type of water bodies in the region) and mature lakes at the final stage of their development, with a diameter larger than 500 m. For this analysis, we selected the major components of the lake water (DOC, pH, Ca, K), low-soluble trace metals (Al, Zr), several environmentally important heavy metals, such as Mn, Co, Cu, Ni, Pb and Cr, and soluble elements bearing the signature of marine deposits or aerosols (Sr, Mo). There are four distinct patterns of element concentrations in the lake water as a function of lake position on the latitude profile that is illustrated in Fig. 12a–l: (1) A decrease in element concentration from the south to the north with a factor of 2 to 4 (DOC, Pb, V, Ga, Ba, Sb), (2) nonsystematic evolution of concentration with a relatively small variation in the latitude with less than a factor of 2 decrease from south to north (Si, Fe, Al, As, REEs), (3) a maximum concentration at the northern border of the discontinuous permafrost site in the continental zone of the N Urengoy site, up to factor of 5 compared with adjacent sites and a factor of 2 to 3 decrease to the northern site sites (Mn, Co, Cd, Cs, Zr, Hf, Th); and (4) an increase in concentration from the south to the north, notably in the coastal Gyda zone, with a factor of 3 to 10, depending on the element and the size of the water body (specific conductivity, Cl, SO_4, Ca, Na, Mg, K, Li, B, Sr, Rb, Mo, W, U, Zn, Cu, Ni and Cr). Although based on sufficient statistics (> 100 lakes, ponds and depressions across the permafrost-bearing zone), this classification should be considered a first order approximation of element behavior along the latitudinal profile, given that (i) depending on the water body size range, the degree of element concentration changes may vary by ±100 % from the values given above, and (ii) the same element (such as Sb) may belong to more than one group of TEs with overall concentration decreases with a local maximum at the Novyi Urengoy site.

The latitudinal profile of DOC concentration, which was based on average values of different size thermokarst lakes of Western Siberia, from discontinuous/sporadic to continuous permafrost zone, demonstrates a general decrease [DOC], more than three times from the initial stage (< 10 m lake) to mature lakes of > 500 m diameter (Fig. 12a). For example, within the sequence of lake size/stage of development, in the sporadic permafrost abundance of the northern taiga zone, the DOC concentration decreases from 36.6 ± 12.6 to $12.7\,\mathrm{mg\,L^{-1}}$, in the forest-tundra zone of discontinuous permafrost this decrease is from 43.9 ± 34.7 to $11 \pm 2.8\,\mathrm{mg\,L^{-1}}$ and in the tundra zone of continuous permafrost it decreases from 19.7 ± 1.5 to $4.4 \pm 0.5\,\mathrm{mg\,L^{-1}}$. It is therefore possible that the increase of thermokarst thawing in the north of western Siberia will increase the concentration of DOC in small size surface water bodies to as much as 300 %. This observation may be due to the enhanced mobilization of organic carbon from peat deposits at elevated temperatures and to an increase in the duration of the active period. In contrast, large size (mature) lakes that are subjected to draining are likely to decrease the DOC concentration, which may be linked to the decrease of the thickness of peat layer deposits feeding the lakes within the transect from the south to the north.

The effect of atmospheric local and long-range input can be accessed via the comparison of dissolved element concentrations in otherwise similar thermokarst lakes that are located at different distances from the sea or from the industrial pollution center. It can be seen that, with the approach to the sea (northward), the concentration of Ca^{2+} and major cations and anions as well as typical trace elements that are enriched in the seawater relative to the continental waters, such as Sr (Fig. 12m) and Mo (Fig. 12n), increases for the lakes of all size ranges. Concerning the heavy metal behavior, there is a pronounced increase of Cu, Ni and Cr concentrations (Fig. 12i, k and l, respectively) in the most northern region of the continuous permafrost zone (Gyda site) within a distance of approximately 300 km from the largest arctic smelter (Cu-Ni-Cr processing factories in Norilsk). This increase is clearly observed for large lakes, which are more capable of integrating the atmospheric deposition. For the other heavy metals, the effect of the proximity to the sea or to the industrial center is not clearly pronounced. It is noteworthy that the concentration of Pb decreases northward, with the change of permafrost regime from discontinuous/sporadic to continuous (Fig. 12j). This decrease may reflect the decrease of the intensity of Pb leaching from peat, which is known to accumulate this element (Shotyk et al., 2000). This decrease of leaching intensity is due to the decrease of the active (unfrozen) layer depth that can supply the peat and related elements to the lake water, as also discussed in the section "Correlations between elements in the thaw lake water bodies".

It follows that, with progressive heating of western Siberian soils, the thawing of the frozen peat, the increasing depth of the active layer and moving the tree line northward, the concentration of most insoluble trace metals that are bound to organic and organo-ferric colloids will increase by a factor of less than 2. The elements that are enriched in peat, such as Pb, V, and Ba, may increase their concentration in thermokarst lakes by a factor of 2 to 3 if the depth of unfrozen peat deposit feeding thaw lakes increases. Soluble elements, such as alkali and alkaline-earth and elements bearing the signature of marine deposits (Li, B, Sr, Rb, Mo, W, U), are likely to remain conservative, given that their maximal concentration is observed already at the most northern continuous permafrost site. An increase by a factor of 2 for DOC and most TE concentrations are demonstrated to occur due to water warming and prolonged summer drought in western Siberia (Pokrovsky et al., 2013), which may further accentuate the organic and organo-mineral colloids and insoluble element transports from the soils to the ocean via lakes and rivers.

4 Conclusions

Toward a better understanding of chemical composition of thermokarst (thaw) lake waters in the subarctic, this work revealed general features of thaw depressions, ponds and lakes in the continental part of the discontinuous and coastal region of the continuous permafrost zone of the northern part of western Siberia. Within the discontinuous permafrost zone of thermokarst lakes, we observe a systematic evolution of water body chemical composition during water ecosystem development, from small permafrost subsidences and depressions and thaw ponds toward large size thermokarst lakes. This trend consists of decreasing DOC, Fe and many insoluble chemical element concentrations during ecosystem maturation and lake size increase, which is most likely linked to peat leaching at the lake border and heterotrophic DOM transpiration in the water column. After lake drainage and active macrophyte growth in the khasyreys (drained lakes), the DOC and some elements, such as K, Rb, Cu, Mg, B, and Sb, increase again due to active autochthonous biological processes.

Compared with "continental" lakes that are located at the watershed divide of the discontinuous permafrost zone, the coastal thermokarst lakes of continuous permafrost exhibit a factor of 2 to 10 lower concentration of DOC and most insoluble elements (Al, Fe, trivalent and tetravalent hydrolysates), a factor of 2 to 5 higher concentration of anions, alkali and alkaline-earth elements, and these lakes do not exhibit any clear tendency in the majority of dissolved constituent chemical compositions as a function of lake size, which is strongly influenced by the proximity to the sea. This difference is important for the extrapolation of the obtained hydrochemical data to larger territories of subarctic water bodies, given that the majority of available information on thermokarst lakes in other subarctic regions belongs to the coastal zone.

Based on the current state of permafrost rocks in the north of western Siberia, one can foresee the increase in the intensity of thermokarst processes on this territory (Kirpotin et al., 2009; Bryksina et al., 2009; Bryskina, 2011; Dneprovskaya et al., 2009; Bryksina and Kirpotin, 2012). This increase should lead to an increase in the number of small soil subsidences and permafrost depressions, together with the decrease in the number of large (mature) lakes. As a result, one can expect an increase in the concentration of dissolved, but low, bio-available soil organic matter and most trace metals that are present as organic and organo-mineral colloids. Based on empirical dependencies between the size (stage development) and concentrations of dissolved ($< 0.22\,\mu$m) trace elements, the average concentration increase may range from 200 to 400 %. Further drainage of these water bodies to the river networks may bring about significant increases in OC, metal nutrients and insoluble elements that are delivered from the wetland zone to the Arctic Ocean.

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Table 1. Physical and major chemical parameters of studied lakes.

Coordinates	Sample name	Stages of evolution	Diameter of the water body, m	pH	EC, µS cm^{-1}	DOC, ppm	Cl$^-$, ppm	SO$_4^{2-}$, ppm	UV 280$_{nm}$
65°59′32.9″/77°51′23.1″	RM1	2nd stage	32	4.29	13.3	14.8	0.15	0.63	0.29
65°59′34.6″/77°51′13.4″	RM2	2nd stage	90	4.73	6.1	3.67	0.059	0.63	0.03
65°59′34.5″/77°51′07.4″	RM3	1st stage	6	4.82	8	10.7	0.091	0.23	0.21
65°59′40.6″/77°51′10.3″	RM4	1st stage *	8	6.92	3.1	4.14	0.061	0.31	0.048
65°59′42.8″/77°51′09.0’	RM5	1st stage *	7	4.74	5.5	8.21	0.04	0.048	0.1
65°59′42.8″/77°51′04.3″	RM6	1st stage *	5	4.6	5.4	6.05	0.035	0.18	0.077
65°59′43.0″/77°51′00.6″	RM7	1st stage *	3	4.73	6.3	5.82	0.051	0.16	0.12
65°59′42.4″/77°50′49.7″	RM8	1st stage	6	4.38	14	21	0.079	0.058	0.41
65°59′39.6″/77°50′22.8″	RM9	3rd stage	123	4.83	6.8	4.31	0.12	0.62	0.08
65°59′49.3″/77°51′25.3″	RM10	1st stage	10	4.23	14	9.59	0.13	1.06	0.2
65°59′49.3″/77°52′09.2″	RM11	4th stage	507	5.35	4.6	4.05	0.074	0.46	0.047
66°00′33.9″/74°45′42.3″	RM13	1st stage	8	3.97	32.9	36.4	0.041	0.062	1.25
66°00′33.8″/74°45′39.2″	RM14	1st stage	9	3.91	29.2	31.5	0.016	0.066	0.89
66°00′34.0″/74°45′34.9″	RM15	1st stage	10	4.03	27.2	47.9	0.044	0.038	0.96
66°00′34.9″/74°45′28.0″	RM16	3rd stage	445	4.85	14.9	16.2	0.14	0.48	0.57
66°01′01.2″/74°45′21.4″	RM17	4th stage	520	4.5	14.8	15.6	0.16	0.51	0.51
65°55′31.8″/74°46′41.1″	RM18	5th stage	260	4.45	12.5	14.2	0.018	0.26	0.34
65°55′28.8″/74°46′45.8″	RM19	5th stage	40	4.93	12.2	13.9	0.02	0.16	0.36
65°55′16.1″/74°49′02.7″	RM20	4th stage	1080	4.98	17.6	8.62	0.2	3.51	0.12
65°52′10.5″/74°52′46.4″	RM21	2nd stage	94	4.43	10.9	9.34	0.1	0.42	0.29
65°54′09.9″/74°08′57.2″	RM22	5th stage	60	4.54	12.2	16.9	0.031	0.13	0.44
65°53′07.0″/75°10′37.7″	RM23	4th stage	810	5.01	11.6	9.75	0.3	0.89	0.19
65°53′29.8″/75°11′37.7″	RM24	4th stage	860	4.85	10	12.5	0.13	0.34	0.27
65°53′58.2″/75°12′38.2″	RM25	4th stage	1100	4.93	12.4	13.1	0.24	0.96	0.26
65°54′15.0″/75°12′24.0″	RM26	5th stage	147	4.8	8.8	13.6	0.02	0.15	0.31
65°51′55.7″/75°20′22.7″	RM27	5th stage	110	4.8	11.4	17.2	0.027	0.24	0.4
65°51′51.5″/75°20′19.1″	RM28	5th stage	54	5.05	11.4	17.3	0.089	0.05	0.41
65°51′59.5″/75°16′49.9″	RM29	5th stage	15	4.34	21.6	21.4	0.021	0.26	0.61
65°51′57.8″/75°16′37.0″	RM30	5th stage	85	4.49	15.6	18.5	0.094	0.41	0.45

Table 1. Continued.

Coordinates	Sample name	Stages of evolution	Diameter of the water body, m	pH	EC, µS cm^{-1}	DOC, ppm	Cl^{-}, ppm	SO$_4^{2-}$, ppm	UV 280$_{nm}$
65°52′44.9″/74°57′42.7″	RM31	3rd stage	490	4.84	9.1	11.4	0.12	0.32	0.29
65°47′47.1″/75°27′08.1″	RM32	4th stage	1200	4.87	8.9	10.2	0.14	0.33	0.22
65°47′22.8″/75°28′31.0″	RM33	2nd stage *	86	6.42	10.4	13.9	0.054	0.48	0.35
65°46′47.4″/75°28′11.7″	RM34	3rd stage	140	5.14	11.2	10.8	0.16	0.11	0.24
65°46′39.8″/75°27′34.9″	RM35	5th stage	50	4.86	10.9	15.5	0.038	0.045	0.47
65°45′31.8″/75°30′51.4″	RM36	5th stage	56	4.89	7.3	7.88	0.026	0.18	0.16
65°45′00.0″/75°31′07.4″	RM37	4th stage	540	5.22	12.6	12.4	0.34	1.09	0.23
65°44′11.0″/75°32′57.0″	RM38	2nd stage	55	4.43	15.4	14.8	0.016	0.24	0.41
65°44′18.5″/75°33′20.0″	RM39	2nd stage	42	3.9	35.8	27.8	0.027	0.61	0.85
65°44′23.5″/75°33′21.8″	RM40	4th stage	1120	4.66	13.3	13.5	0.18	1.004	0.34
66°00′58.6″/74°47′42.5″	RM41	2nd stage	54	4.29	18.2	8.73	0.022	0.17	0.56
66°01′01.6″/74°47′45.5″	RM42	2nd stage	88	4.12	26.3	27.3	0.027	0.17	0.82
66°01′18.2″/74°47′32.1″	RM43	3rd stage	154	4.71	11.1	12.4	0.076	0.28	0.37
66°00′28.9″/74°48′30.7″	RM44	2nd stage	62	4.41	18.5	17.3	0.014	0.12	0.53
70°50′42.7″/78°33′33.8″	RM46	arctic lake	525	6.52	57.7	4.92	3.85	0.46	0.076
	RM47	arctic lake	400	6.35	63	5.36	3.04	0.64	0.101
70°53′43.8″/78°22′7.5″	RM48	arctic lake	150	6.18	40.7	5.15	1.01	0.042	0.011
70°51′4.3″/78°33′16.2″	RM49	arctic lake	440	6.58	60.6	8.18	6.94	0.22	0.096
70°51′31.3″/78°33′8.1″	RM50	arctic lake	12	6.46	64	18.2	4.14	0.2	0.072
70°51′49.3″/78°32′44.4″	RM51	arctic lake	50	6.22	253	21.3	4.13	1.62	0.54
70°51′51.9″/78°30′18.1″	RM52	arctic lake	295	6.42	59.5	4.2	1.51	0.22	0.34
70°52′42.2″/78°28′7.9″	RM53	arctic lake	700	6.42	66.5	3.81	1.96	0.082	0.078
70°53′9.3″/78°28′11.3″	RM54	arctic lake	365	6.3	76.5	3.82	6.65	0.42	0.037
70°53′29.9″/78°22′14″	RM55	arctic lake	203	6.19	43.5	3.21	1.64	0.16	0.043
70°53′19.6″/78°22′8.2″	RM56	arctic lake	305	6.21	42.1	4	1.87	0.016	0.049
70°53′16.3″/78°22′41.8″	RM57	arctic lake	502	6.5	115	4.35	1.9	0.073	0.055
70°53′16.1″/78°25′0.6″	RM58	arctic lake	130	6.48	43.5	7.54	0.6	0.17	0.12
70°50′44″/78°36′6.9″	RM59	arctic lake	230	6.51	62.2	5.79	3.8	0.6	0.104
70°53′33″/78°30′19.6″	RM60	arctic lake	300	6.45	46.5	5.79	2.22	0.34	0.12

EC – specific conductivity, DOC – dissolved organic carbon, UV 280$_{nm}$ – ultraviolet absorption at a wavelength of 280 nm, "*" – basins and subsidence, filled with low-mineralized rain water which do not reflect the dynamics of the succession of lakes in chemical composition.

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Table 2. Average (±2σ) and the range of element concentration in thermokarst lake waters of different stages from the Novyi Urengoy region (1st–5th stage) and from Gyda district (coastal lakes).

Element	units	1st stage (2–10 m) $n = 6$	2nd stage (11–100 m) $n = 8$	3rd stage (101–500 m) $n = 5$	4th stage (> 500 m) $n = 9$	5th stage (Khasyrey) $n = 10$	Arctic coastal Lakes (Gyda) $n = 15$	Average river water concentration*
Na	mg L^{-1}	0.05–0.72 0.3 ± 0.25	0.021–0.32 0.14 ± 0.12	0.365–0.433 0.4 ± 0.029	0.206–0.8 0.47 ± 0.21	0.055–0.246 0.17 ± 0.069	0.76–14.04 2.62 ± 3.33	–
Mg	mg L^{-1}	0.14–0.386 0.24 ± 0.1	0.072–0.266 0.19 ± 0.06	0.143–0.391 0.24 ± 0.097	0.072–0.472 0.3 ± 0.12	0.194–0.535 0.35 ± 0.11	0.819–2.71 1.48 ± 0.51	–
Al	mg L^{-1}	0.075–0.262 0.16 ± 0.075	0.04–0.198 0.11 ± 0.06	0.032–0.13 0.086 ± 0.049	0.043–0.149 0.081 ± 0.031	0.047–0.187 0.11 ± 0.047	0.002–0.061 0.015 ± 0.018	0.032
Si	mg L^{-1}	0.2–1.99 0.85 ± 0.7	0.175–0.943 0.3 ± 0.26	0.184–0.349 0.26 ± 0.061	0.167–0.355 0.23 ± 0.067	0.172–0.825 0.37 ± 0.21	0.007–1.16 0.23 ± 0.33	–
K	mg L^{-1}	0.01–0.062 0.031 ± 0.017	0.011–0.064 0.042 ± 0.019	0.067–0.138 0.11 ± 0.03	0.049–0.27 0.14 ± 0.065	0.004–0.119 0.03 ± 0.034	0.084–0.855 0.33 ± 0.21	–
Ca	mg L^{-1}	0.238–0.93 0.49 ± 0.25	0.125–0.67 0.4 ± 0.16	0.205–0.668 0.44 ± 0.17	0.125–0.879 0.54 ± 0.22	0.362–0.756 0.53 ± 0.14	0.442–1.5 0.85 ± 0.3	–
Fe	mg L^{-1}	0.12–2.36 1.2 ± 1	0.031–0.705 0.38 ± 0.25	0.057–0.382 0.22 ± 0.15	0.03–0.297 0.13 ± 0.079	0.152–0.742 0.36 ± 0.21	0.024–0.91 0.17 ± 0.23	0.066
Li	µg L^{-1}	0.385–1.05 0.65 ± 0.25	0.149–0.963 0.49 ± 0.27	0.287–1.03 0.69 ± 0.29	0.143–1.358 0.81 ± 0.34	0.715–1.84 1.07 ± 0.36	–	1.84
B	µg L^{-1}	1.76–2.62 2.1 ± 0.33	1.74–2.64 2.1 ± 0.33	1.35–4.18 2.2 ± 1.1	1.28–4.92 2.8 ± 1.03	0.837–2.94 2.04 ± 0.73	0.711–6.97 3.1 ± 2	10.2
Ti	µg L^{-1}	2.95–4.57 3.8 ± 0.7	2.47–3.74 3.2 ± 0.5	2.92–4.71 3.6 ± 0.82	2.26–4.51 3.03 ± 0.83	2.27–3.33 2.8 ± 0.35	2.21–5.71 3.4 ± 0.79	0.489
V	µg L^{-1}	0.181–0.771 0.35 ± 0.2	0.118–0.644 0.33 ± 0.19	0.1–0.799 0.45 ± 0.29	0.227–1.1 0.41 ± 0.27	0.113–0.286 0.19 ± 0.061	0.043–0.667 0.2 ± 0.19	0.71
Cr	µg L^{-1}	3.47–5.48 4.3 ± 0.7	2.56–4.04 3.3 ± 0.6	3.12–4.12 3.8 ± 0.4	2.87–4.66 3.5 ± 0.61	2.77–4.11 3.4 ± 0.4	2.77–7.16 4.3 ± 1.1	0.7
Mn	µg L^{-1}	5.44–48.1 20 ± 16	3.53–44.5 17 ± 13	3.1–34.6 14.03 ± 12.4	4.77–34.01 21.8 ± 9.8	6.23–52.1 27.2 ± 13.9	0.375–20.4 5.1 ± 5.2	34
Co	µg L^{-1}	0.319–1.17 0.7 ± 0.3	0.19–0.68 0.43 ± 0.17	0.093–0.715 0.3 ± 0.25	0.169–0.847 0.48 ± 0.24	0.155–0.963 0.6 ± 0.22	0.002–0.439 0.08 ± 0.11	0.148
Ni	µg L^{-1}	2.48–4.26 3.4 ± 0.6	1.87–2.8 2.4 ± 0.3	2.38–3.09 2.8 ± 0.27	1.89–3.21 2.6 ± 0.47	2.26–3.42 2.7 ± 0.36	2.09–5.63 3.3 ± 0.91	0.801

Table 2. Continued.

Element	units	1st stage (2–10 m) *n* = 6	2nd stage (11–100 m) *n* = 8	3rd stage (101–500 m) *n* = 5	4th stage (> 500 m) *n* = 9	5th stage (Khasyrey) *n* = 10	Arctic coastal Lakes (Gyda) *n* = 15	Average river water concentration*
Cu	µg L^{-1}	0.161–0.487 0.3 ± 0.13	0.085–0.245 0.17 ± 0.06	0.161–0.49 0.34 ± 0.12	0.086–0.664 0.48 ± 0.18	0.104–0.427 0.25 ± 0.1	0.298–11.6 1.6 ± 2.8	1.48
Zn	µg L^{-1}	7.28–63.9 22.6 ± 22.7	3.94–13.6 8.2 ± 3	2.3–8.3 5.6 ± 2.6	3.37–195.1 26.5 ± 63.3	3.46–20.5 10.2 ± 5	1.38–37.5 8.5 ± 10.3	0.6
Ga	µg L^{-1}	0.021–0.044 0.029 ± 0.009	0.017–0.048 0.03 ± 0.01	0.0009–0.015 0.006 ± 0.006	0.0002–0.01 0.003 ± 0.002	0.0001–0.012 0.004 ± 0.004	0.0002–0.009 0.004 ± 0.003	0.03
As	µg L^{-1}	0.37–1.33 0.9 ± 0.3	0.406–0.806 0.63 ± 0.13	0.28–0.841 0.57 ± 0.2	0.313–0.777 0.55 ± 0.15	0.408–0.931 0.63 ± 0.17	0.249–0.635 0.38 ± 0.11	0.62
Se	µg L^{-1}	0.17–0.31 0.23 ± 0.05	0.202–0.273 0.23 ± 0.02	0.37–0.48 0.4 ± 0.04	0.33–0.6 0.41 ± 0.077	0.37–0.48 0.41 ± 0.03	–	0.07
Rb	µg L^{-1}	0.041–0.242 0.12 ± 0.07	0.053–0.275 0.16 ± 0.08	0.217–0.577 0.32 ± 0.16	0.15–0.684 0.28 ± 0.16	0.037–0.275 0.1 ± 0.07	0.097–0.549 0.29 ± 0.15	1.63
Sr	µg L^{-1}	2.74–5.75 4.7 ± 1.2	1.3–4.17 3.1 ± 0.9	2.15–4.31 3.3 ± 0.9	1.33–9.55 4.5 ± 2.3	2.62–5.102 4.1 ± 0.84	4.42–12.8 7.7 ± 2.5	60
Y	µg L^{-1}	0.032–0.176 0.08 ± 0.05	0.018–0.067 0.044 ± 0.016	0.024–0.1 0.056 ± 0.029	0.019–0.122 0.072 ± 0.037	0.035–0.139 0.063 ± 0.033	–	0.04
Zr	µg L^{-1}	0.142–0.702 0.32 ± 0.2	0.094–0.244 0.16 ± 0.05	0.112–0.27 0.19 ± 0.062	0.09–0.33 0.24 ± 0.073	0.114–0.3 0.2 ± 0.061	0.002–0.214 0.033 ± 0.056	0.039
Mo	µg L^{-1}	0.09–0.107 0.098 ± 0.006	0.082–0.099 0.091 ± 0.006	0.069–0.089 0.077 ± 0.007	0.055–0.095 0.071 ± 0.012	0.055–0.076 0.062 ± 0.006	0.03–0.1 0.059 ± 0.023	0.42
Cd	µg L^{-1}	0.079–0.136 0.1 ± 0.02	0.074–0.099 0.087 ± 0.009	0.028–0.035 0.032 ± 0.003	0.021–0.04 0.03 ± 0.006	0.027–0.044 0.037 ± 0.005	0.015–0.035 0.023 ± 0.006	0.08
Sb	µg L^{-1}	0.044–0.07 0.06 ± 0.01	0.049–0.073 0.062 ± 0.009	0.045–0.059 0.054 ± 0.006	0.051–0.112 0.084 ± 0.023	0.04–0.055 0.047 ± 0.005	0.007–0.023 0.013 ± 0.005	0.07
Cs	µg L^{-1}	0.017–0.023 0.019 ± 0.002	0.018–0.022 0.019 ± 0.001	0.012–0.018 0.014 ± 0.003	0.011–0.017 0.012 ± 0.002	0.011–0.014 0.012 ± 0.0008	0.0003–0.003 0.002 ± 0.0009	0.011
Ba	µg L^{-1}	1.43–9.32 4.8 ± 2.9	0.316–4.34 1.8 ± 1.5	1.9–4.8 3.2 ± 1.1	1.6–14.1 4.3 ± 3.8	1.55–5.79 3.1 ± 1.5	0.246–2.65 0.93 ± 0.7	23
La	µg L^{-1}	0.019–0.106 0.05 ± 0.03	0.01–0.045 0.025 ± 0.012	0.019–0.082 0.042 ± 0.025	0.013–0.093 0.051 ± 0.028	0.021–0.113 0.041 ± 0.029	0.005–0.118 0.038 ± 0.037	0.012
Ce	µg L^{-1}	0.048–0.33 0.15 ± 0.1	0.031–0.118 0.074 ± 0.03	0.043–0.213 0.11 ± 0.066	0.031–0.258 0.14 ± 0.078	0.058–0.298 0.11 ± 0.075	0.01–0.267 0.075 ± 0.079	0.262
Pr	µg L^{-1}	0.006–0.042 0.02 ± 0.01	0.004–0.014 0.009 ± 0.004	0.007–0.027 0.015 ± 0.008	0.005–0.033 0.019 ± 0.009	0.009–0.037 0.016 ± 0.009	0.002–0.037 0.011 ± 0.011	0.04
Nd	µg L^{-1}	0.036–0.186 0.09 ± 0.05	0.026–0.07 0.05 ± 0.014	0.026–0.112 0.058 ± 0.034	0.019–0.134 0.076 ± 0.039	0.036–0.148 0.064 ± 0.035	0.006–0.2 0.055 ± 0.061	0.152

Table 2. Continued.

Element	units	1st stage (2–10 m) $n = 6$	2nd stage (11–100 m) $n = 8$	3rd stage (101–500 m) $n = 5$	4th stage (> 500 m) $n = 9$	5th stage (Khasyrey) $n = 10$	Arctic coastal Lakes (Gyda) $n = 15$	Average river water concentration*
Sm	$\mu g\,L^{-1}$	0.011–0.044 0.02 ± 0.01	0.008–0.019 0.014 ± 0.003	0.007–0.026 0.014 ± 0.008	0.005–0.033 0.019 ± 0.009	0.009–0.032 0.015 ± 0.007	0.002–0.048 0.012 ± 0.014	0.036
Eu	$\mu g\,L^{-1}$	0.01–0.018 0.013 ± 0.003	0.007–0.011 0.01 ± 0.001	0.006–0.013 0.009 ± 0.002	0.006–0.015 0.011 ± 0.003	0.007–0.013 0.009 ± 0.002	0.0002–0.01 0.003 ± 0.003	0.0098
Gd	$\mu g\,L^{-1}$	0.013–0.049 0.03 ± 0.01	0.011–0.022 0.017 ± 0.004	0.011–0.033 0.02 ± 0.008	0.01–0.039 0.025 ± 0.011	0.015–0.041 0.021 ± 0.008	0.002–0.056 0.014 ± 0.015	0.04
Tb	$\mu g\,L^{-1}$	0.003–0.008 0.005 ± 0.002	0.003–0.005 0.004 ± 0.0004	0.0003–0.003 0.002 ± 0.001	0.0001–0.004 0.002 ± 0.001	0.0009–0.004 0.002 ± 0.001	–	0.0055
Dy	$\mu g\,L^{-1}$	0.008–0.034 0.02 ± 0.009	0.005–0.015 0.01 ± 0.003	0.006–0.021 0.012 ± 0.006	0.005–0.025 0.016 ± 0.008	0.007–0.025 0.012 ± 0.006	0.002–0.042 0.01 ± 0.011	0.03
Ho	$\mu g\,L^{-1}$	0.005–0.01 0.007 ± 0.002	0.004–0.006 0.005 ± 0.0007	0.003–0.006 0.004 ± 0.001	0.002–0.006 0.005 ± 0.002	0.003–0.007 0.004 ± 0.001	0.0004–0.042 0.003 ± 0.003	0.0071
Er	$\mu g\,L^{-1}$	0.013–0.03 0.02 ± 0.006	0.012–0.017 0.015 ± 0.002	0.004–0.014 0.008 ± 0.004	0.004–0.016 0.01 ± 0.005	0.006–0.017 0.009 ± 0.003	0.001–0.03 0.007 ± 0.007	0.02
Tm	$\mu g\,L^{-1}$	0.007–0.01 0.008 ± 0.001	0.007–0.008 0.0074 ± 0.0004	0.004–0.005 0.0042 ± 0.0004	0.004–0.005 0.004 ± 0.0006	0.004–0.005 0.004 ± 0.0004	0.0003–0.005 0.002 ± 0.001	0.0033
Yb	$\mu g\,L^{-1}$	0.008–0.025 0.01 ± 0.006	0.006–0.012 0.009 ± 0.002	0.005–0.014 0.009 ± 0.003	0.005–0.015 0.011 ± 0.004	0.006–0.016 0.009 ± 0.003	0.0003–0.036 0.007 ± 0.009	0.017
Lu	$\mu g\,L^{-1}$	0.003–0.005 0.004 ± 0.0008	0.003–0.004 0.0034 ± 0.0004	0.002–0.003 0.003 ± 0.0005	0.002–0.004 0.003 ± 0.0006	0.002–0.003 0.003 ± 0.0004	0.001–0.006 0.002 ± 0.001	0.0024
Hf	$\mu g\,L^{-1}$	0.027–0.044 0.03 ± 0.006	0.025–0.03 0.028 ± 0.002	0.025–0.031 0.028 ± 0.002	0.025–0.031 0.029 ± 0.002	0.025–0.031 0.028 ± 0.002	0.0001–0.008 0.003 ± 0.002	0.0059
W	$\mu g\,L^{-1}$	0.058–0.098 0.07 ± 0.02	0.048–0.084 0.063 ± 0.014	0.051–0.086 0.063 ± 0.014	0.037–0.066 0.048 ± 0.01	0.038–0.089 0.056 ± 0.016	0.031–0.25 0.064 ± 0.055	0.1
Pb	$\mu g\,L^{-1}$	0.095–0.512 0.28 ± 0.2	0.028–0.316 0.15 ± 0.097	0.041–0.11 0.079 ± 0.034	0.047–0.23 0.088 ± 0.059	0.044–0.17 0.084 ± 0.04	0.024–0.158 0.055 ± 0.035	0.079
Th	$\mu g\,L^{-1}$	0.01–0.08 0.03 ± 0.02	0.003–0.019 0.01 ± 0.005	0.005–0.025 0.017 ± 0.008	0.004–0.024 0.015 ± 0.006	0.007–0.025 0.015 ± 0.006	0.0001–0.014 0.003 ± 0.004	0.041
U	$\mu g\,L^{-1}$	0.003–0.011 0.005 ± 0.003	0.002–0.007 0.003 ± 0.001	0.003–0.01 0.007 ± 0.003	0.002–0.012 0.008 ± 0.003	0.003–0.005 0.004 ± 0.001	0.001–0.014 0.005 ± 0.004	0.372

The nominator represents the minimal and maximal value and denominator represents the average value with standard deviation; *n* is the number of studied water objects; * stands for (Gaillardet et al., 2003).

Table 3. Pearson correlation coefficients between element concentration and thaw lake size (arctic coastal lakes (Gyda)).

Elements	DOC	pH	Mg	Ca	Fe	Si	K	Na	Al	Mn	Co	Zr	La	Ce	Ho	Yb	Lu	Th
Pearson correlation coefficients	−0.61	0.37	−0.23	−0.11	−0.57	−0.58	0.33	0.09	−0.53	−0.62	−0.60	−0.57	−0.38	−0.40	−0.55	−0.52	−0.55	−0.55

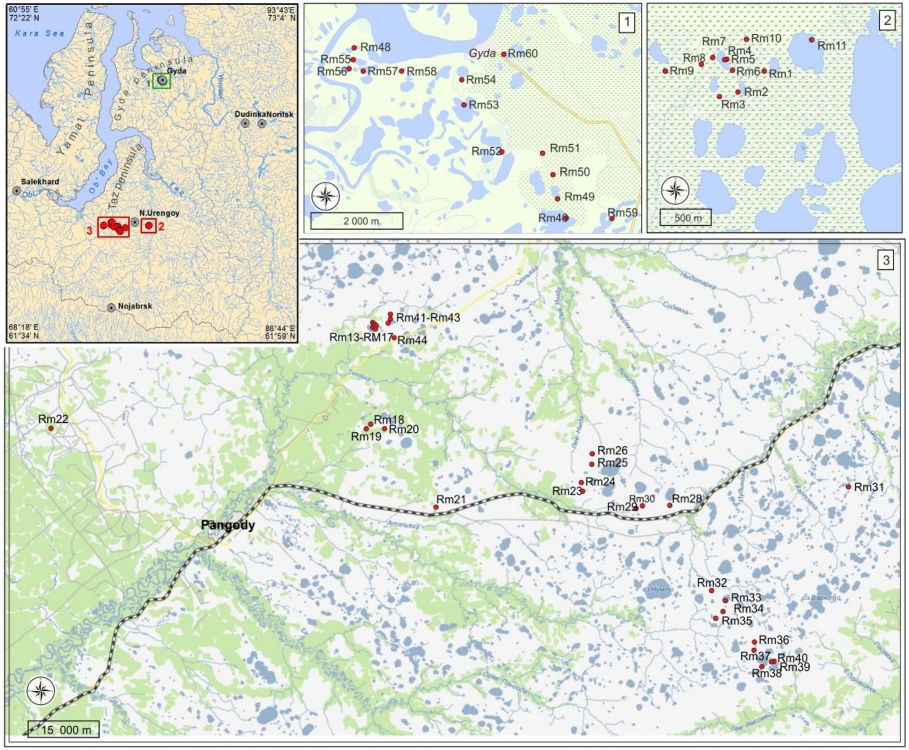

Fig. 1. Map of the studied region. Sites 1, 2 and 3 correspond to the continental zone of discontinuous permafrost, including the district of Pangody (Yamalo–Nenetsk Region), west of the Novyi Urengoy region in August 2010; the east of the Novyi Urengoy region in August 2011, and the coastal zone, around the village of Gyda within the zone of continuous permafrost sampled in August 2011, respectively.

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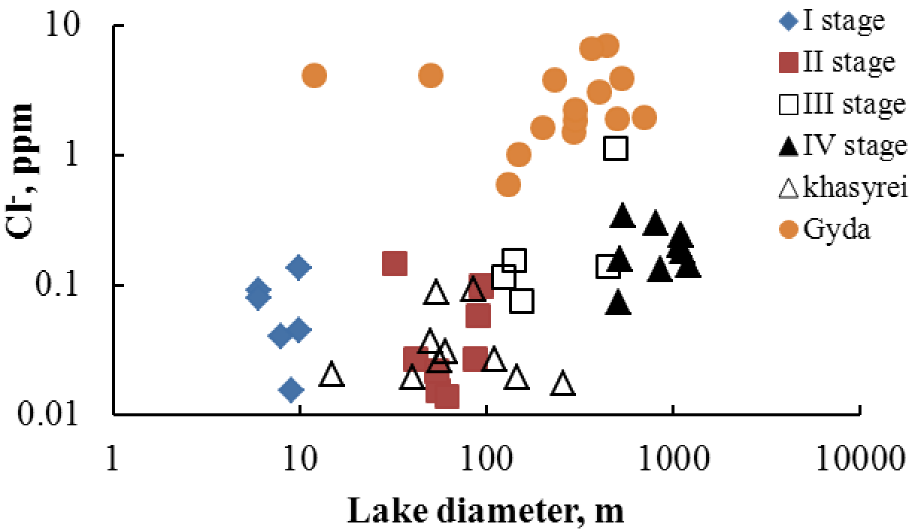

Fig. 2. Aqueous chloride concentration as a function of the lake size in continnetal subractic zone (from the 1st to the 4th stage of formation and khasyrei shown by diamonds, rectangulas and triangules) and continental Arctic region (solid cicrles).

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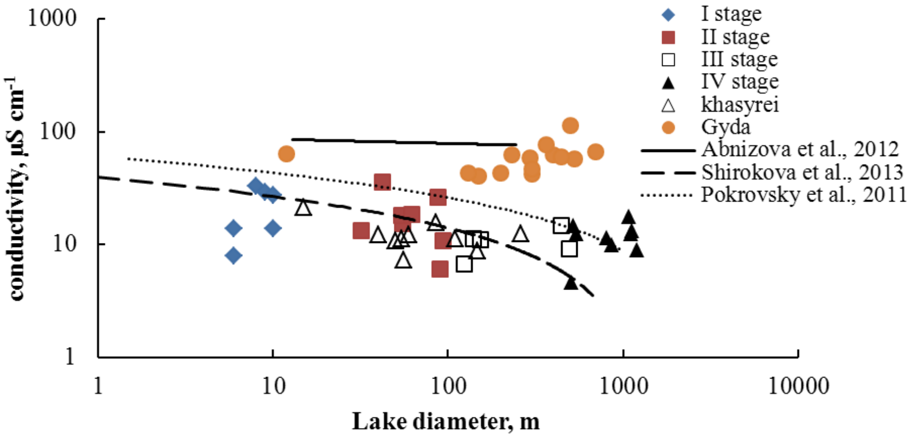

Fig. 3. A plot of lakewater conductivity as a function of lake diameter.

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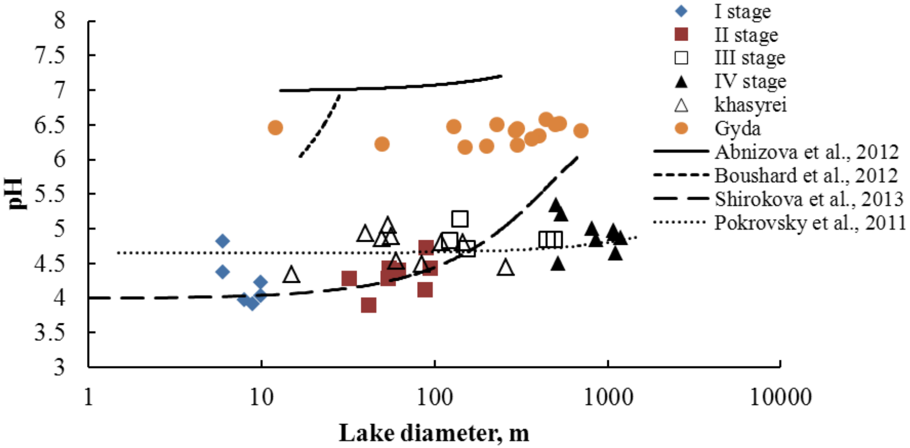

Fig. 4. A plot of lakewater pH as a function of lake diameter.

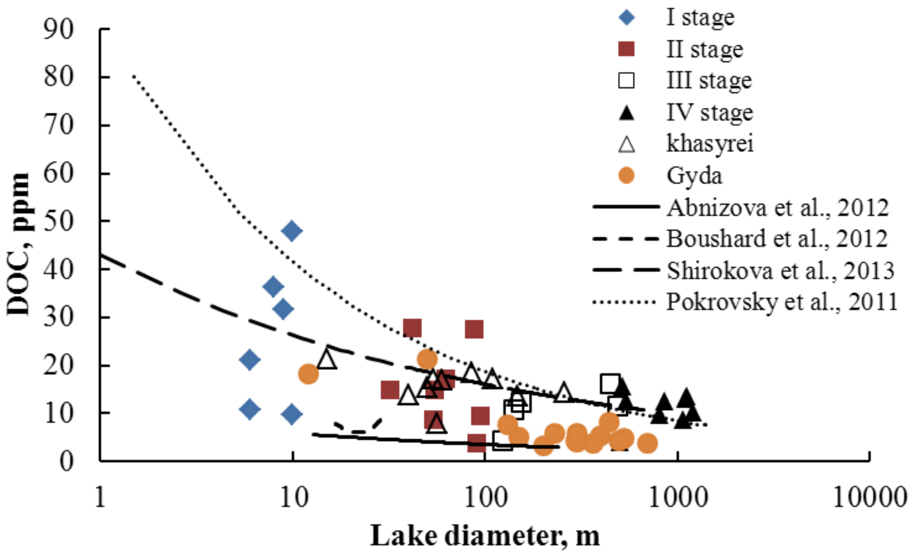

Fig. 5. A plot of DOC (< 0.45 µm) concentration in studied thaw lakes as a function of lake diameter.

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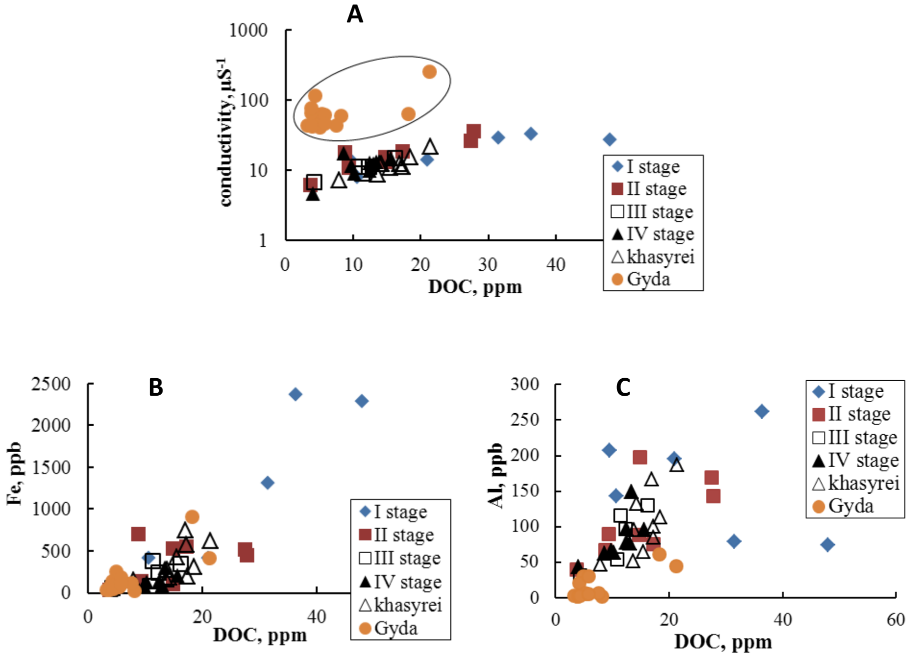

Fig. 6. Dependency of conductivity **(A)**, Fe **(B)** and Al **(C)** concentration on DOC concentration in thermokarst lakes of different stage of development.

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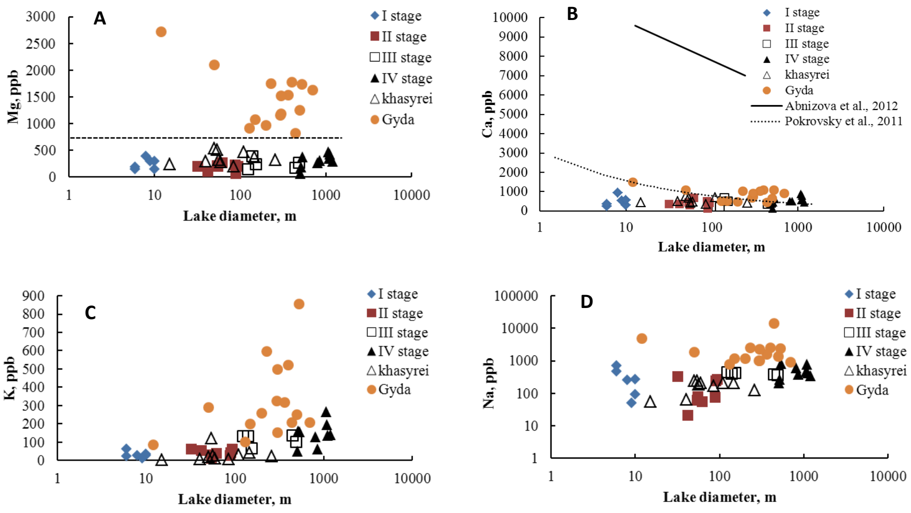

Fig. 7a. Element concentration in the water column as a function of major element concentration: Mg **(A)**, Ca **(B)**, K **(C)**, Na **(D)**.

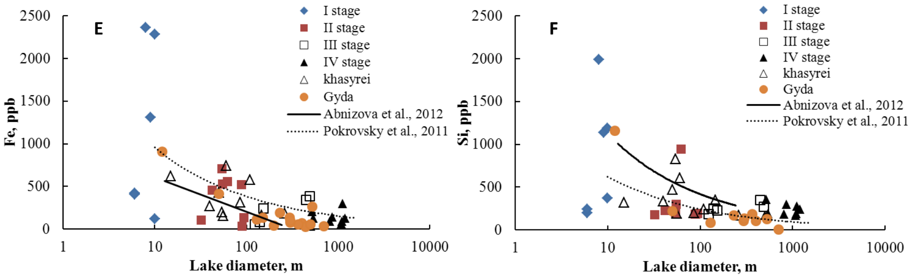

Fig. 7b. Element concentration in the water column as a function of major element concentration: Fe **(E)** and Si **(F)**.

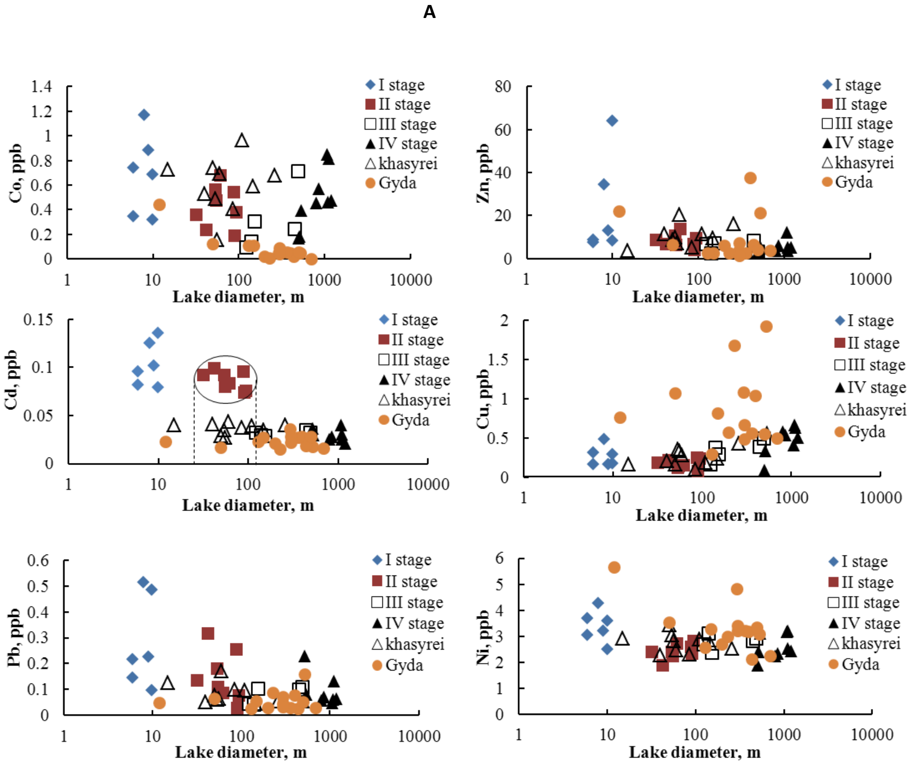

Fig. 8a. Examples of trace elements concentration dependence on the lake size in two studied regions.

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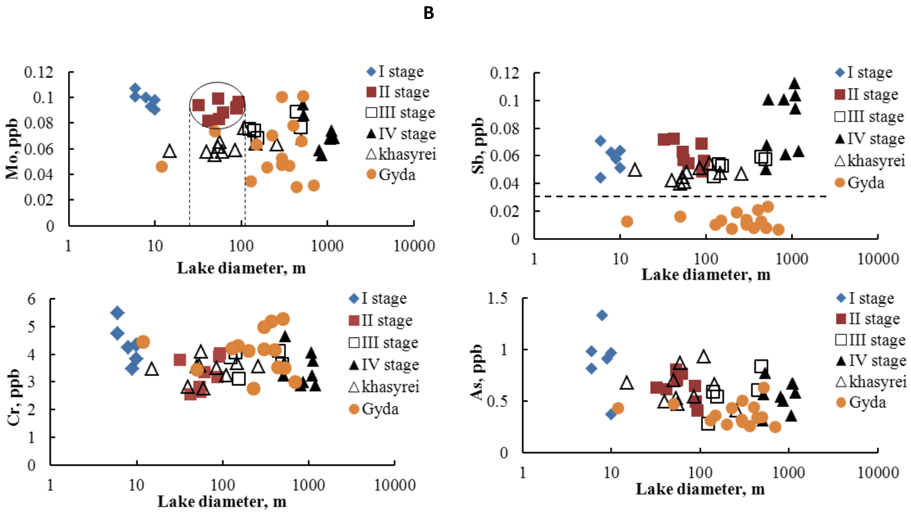

Fig. 8b. Continued.

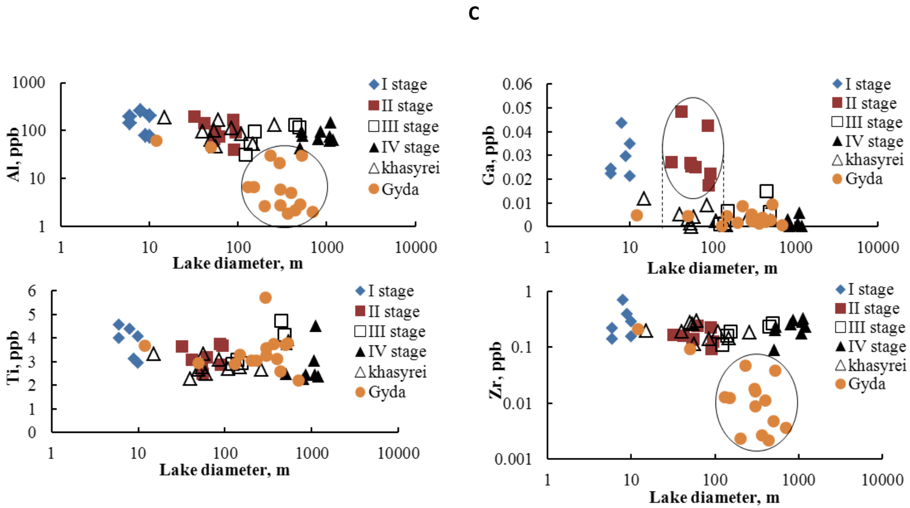

Fig. 8c. Continued.

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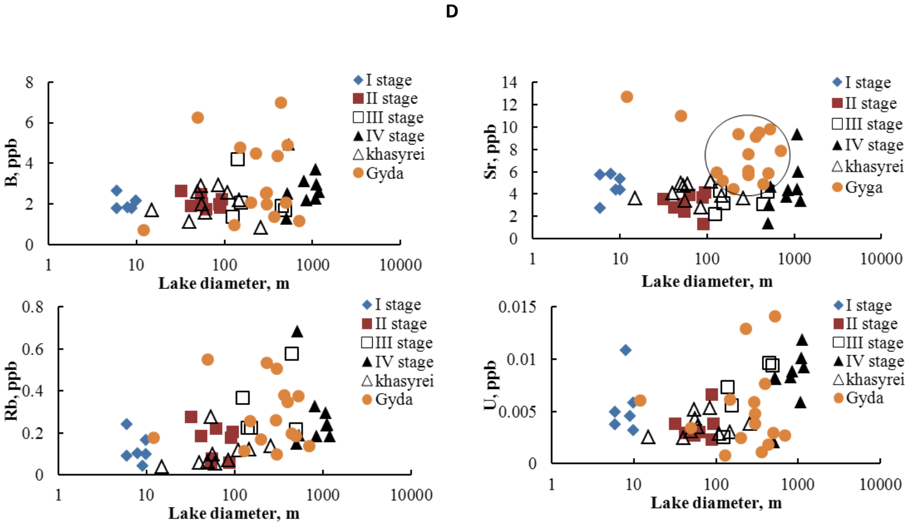

Fig. 8d. Continued.

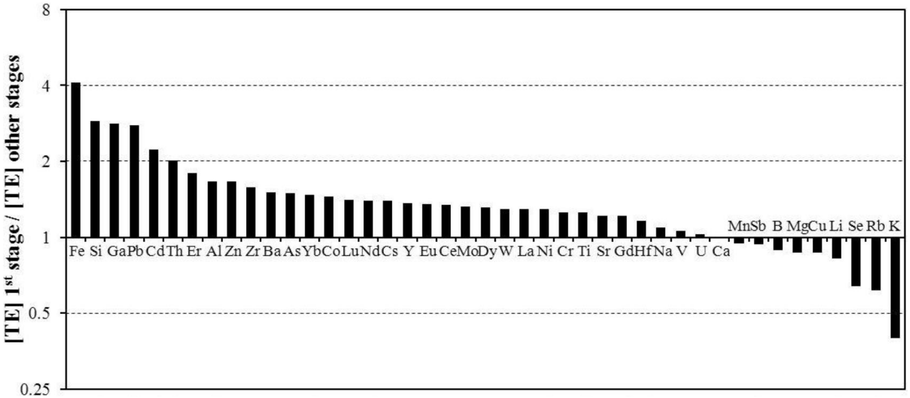

Fig. 9. The ratio of element average concentration in the lakes of 1st stage of formation to the average value of all other stages.

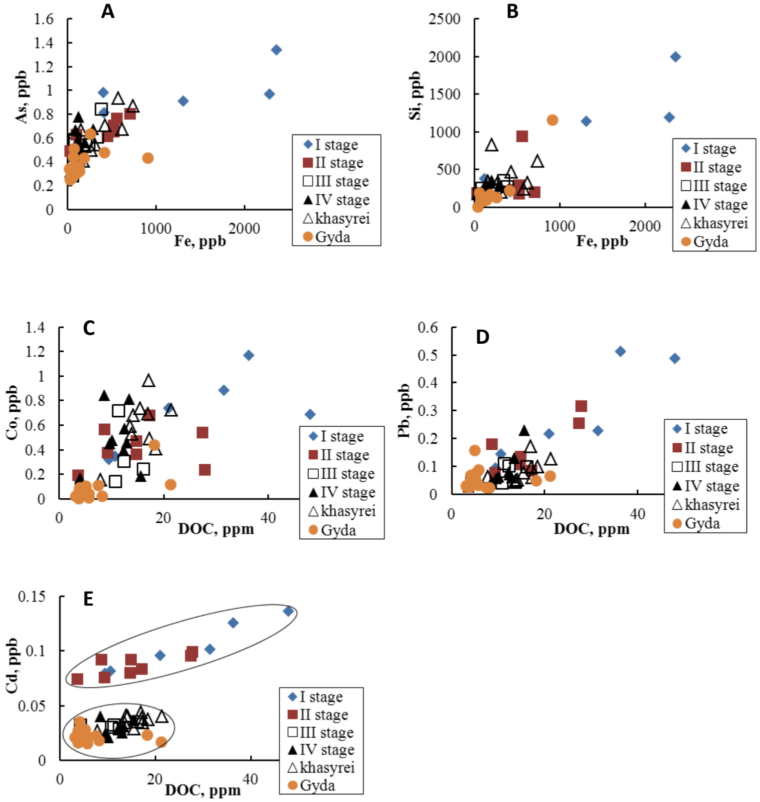

Fig. 10. As **(A)** and Si **(B)** concentration as a function of Fe concentration in the water column of thaw lakes. Correlations between Co **(C)**, Pb **(D)** and Cd **(E)** concentration and that of DOC in thermokarst lakes of two studied regions.

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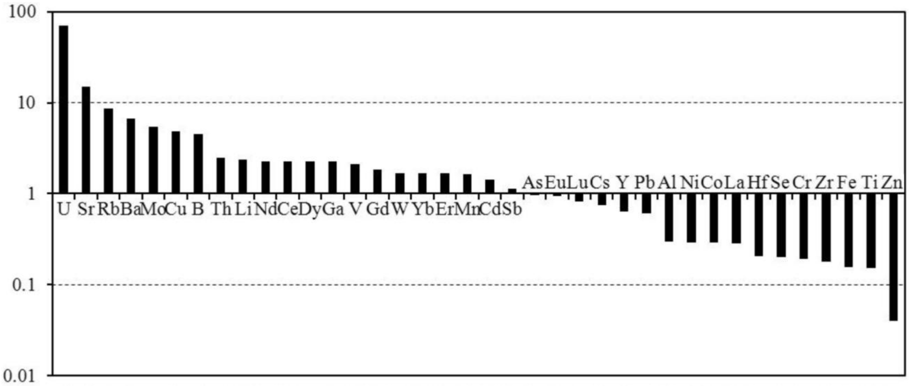

Fig. 11. Concentrations of major and trace elements in two types of studied lake waters and the average river water value (Gaillardet et al., 2003) (log [world average]/[lakes average]).

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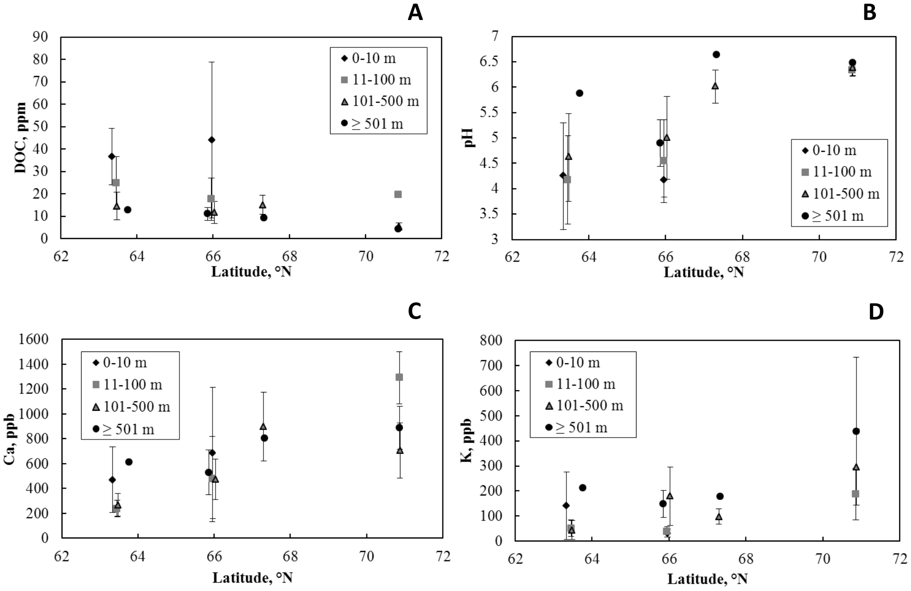

Fig. 12a. The evolution of lake water pH, DOC, Ca, K concentration in thermokarst lakes as a function of latitude in disocntinuous/Sporadic and continuous permafrost zone of western Siberia. The average values with standard deviation are calculated for each stage of lake development, corresponding to different lake diameter. In addition to the results of the present study, data of Shirokova et al., 2009, 2013 and Pokrovsky et al. (2011) were used.

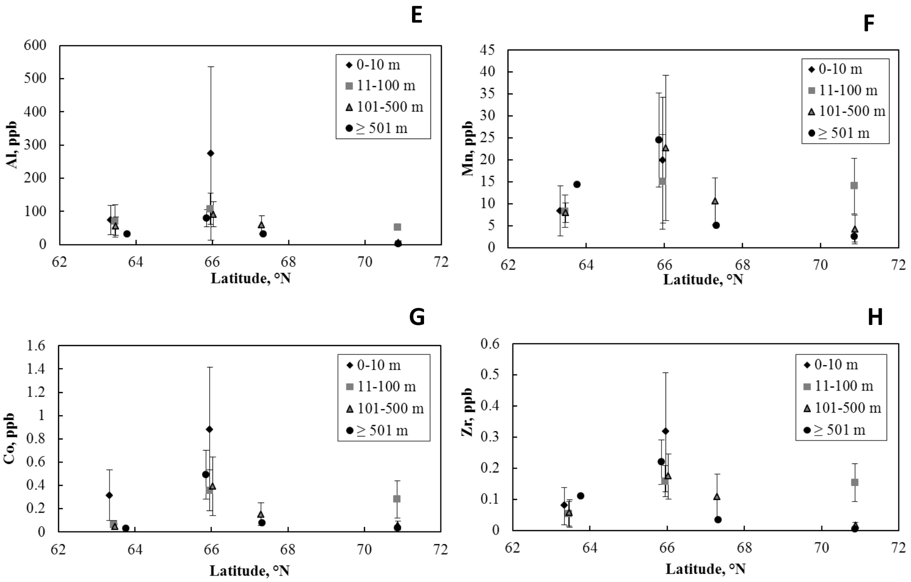

Fig. 12b. Continued.

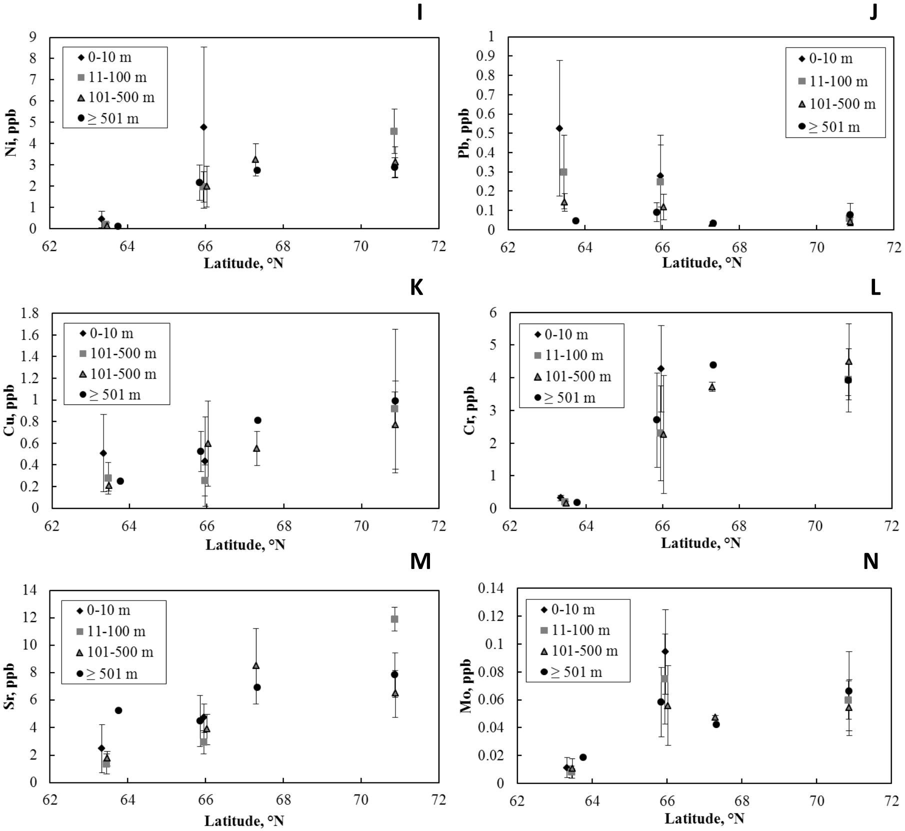

Fig. 12c. Continued.

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