

Responses to Reviewer No 2

The reviewer requested the following modifications:

A) Organization and writing must be improved to increase the focus and clarity of the manuscript; We partially reorganized the introduction and discussion; the text was greatly modified following numerous constructive comments of both reviewers

B) The manuscript needs to be shortened – there are too many figures and sections of the discussion are speculative; We removed 14 figure plots, placed a big deal of other figures in the ESM and shortened the speculative discussion on climate effect as much as the whole manuscript page.

C) The patterns of variation in important chemical parameters or trends in the data should be tested statistically (it is not always clear in the text whether the reported differences or trends are significant). This point is well taken. Element concentrations 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). This is thoroughly described in the end of section 2. In the revised section, we presented all missing statistic parameters to our treatments of element concentration – lake surface area dependence; order of element abundance in the lake waters and element concentration dependence on the latitude.

D) The conclusions should be restricted to points that can be supported by statistical tests or models that describe the nature and significance of the variation in the dataset. This comment is complementary to comment (C) and we carefully addressed the statistical issues in the revised version.

Some specific items which must be considered include:

1) The provision of additional context on the study areas. This remark has been also issued by the first reviewer; in response to this comment we added a comparative table of three sites in the Electronic Annex

2) Provision of a clear explanation of the stages of thermokarst lake development and a justification for using lake size as a criteria for determining the stage of thermokarst lake development, as the latter is used to stratify the water chemistry data set. We added a big deal of explanation / description in section 2.1, as also following the remarks of reviewer No 1

3) The development of refined and concise objectives or hypotheses to focus how the lake dataset will be reported and analyzed, and to provide a framework to focus or constrain the discussion. We reformulated the objectives of this work (end of the Introduction) as also requested by the 1st reviewer

3) The greater use of statistical tests and reporting of test results to describe significant differences in the chemistry of different lake populations or the significance of trends. In the revised version, we extended the description of statistical approaches used in this work and we provided the missing statistical criteria for trends, correlations and differences between samples.

4) Careful review of the text to eliminate editorial errors. We carefully edited the text, figure captions and tables we corrected all errors noted by reviewer; in addition, we provided a number of self-motivated corrections.

I also note that several of the figures require editorial attention and captions have inconsistencies and errors that require correction. We carefully revised these errors and inconsistencies.

Specific comments of reviewer No 2

Title – It would be useful to clarify in the title that the study evaluates lake conditions across a “permafrost gradient” or simply across “permafrost zones”, those being continuous to discontinuous. This comment should also be considered with respect to the running title. It is not clear why “in the context of climate change” is in the title. This can be omitted. We agree and corrected the title as following: “Thermokarst lake waters across permafrost zones of Western Siberia”.

Introduction

I think the Authors should consider providing a few lines of additional context regarding peat thicknesses, nature of underlying mineral sediments, and possibly the mineralogical or geochemical composition of the substrate which characterizes the study region. We agree and added several explicatory sentence in the revised text.

Some additional description of the physical context, including regional physiography, ground ice conditions and nature and rates of thermokarst in the region would help to provide readers with a more clear understanding of the processes affecting these lakes. This would also assist in placing results of this study into a broader regional or global context. In response to this comment, we added a full page of physico-geographical description of the territories in the ESM-1.

Sites 1 and 2 lay within the discontinuous permafrost zone and site 3 is located within continuous permafrost zone. The obtained data were compared with other available information on thermokarst ponds and lakes of western Siberia chemical composition presented elsewhere (Shirokova et al., 2013; Pokrovsky et al., 2013). Thanks to this comparison, we could encompass significant range of climate, vegetation, permafrost thickness and coverage, soil and lithology, from arctic tundra to northern taiga developed on essentially the same mineral and peat substrate. Detailed comparative description of study sites is presented in the Electronic Supporting Information (Table ESM-1) and for convenience given below

Table ESM-1. Physico-geographical characteristics of studied sites of the north of Western Siberia.

Site	MAAT, °C	precipitation	permafrost	vegetation	depth of peat
<u>Gyda No 1</u>	-12	Annual precipitation of 566 mm; ≤ 350 mm in summer; 150–190 mm in winter	Temperature from -5 to -10 C°, depth from 200 to 430 m except at the coast (≤ 200 m)	Tundra (moss, lichens, dwarf shrubs)	0.2 to 0.5 m
<u>Pangody/Novyi Urengoy No 2, 3</u>	-6.9 to -7.6	Annual precipitation 410-460 mm, with 70 to 75 % occurring during snow-free period	Permafrost from 50 to 200 m. Taliks under the thermokarst lakes of 12 to 85 m. Sporadic permafrost around large rivers	Forest-tundra (larch, pine, fir with tree height < 3-5 m. Forest occupies between 20 and 30%, tundra between 5 and	Up to 5.5 m in the river valley and 0.5 to 4 m at the watershed divide

				10%	
<u>Nojabrsk/ Khanymev</u>	-6.5	Annual precipitation of 498 mm (75 – 80% occurs from April to October and 20-25% during cold period)	Sporadic permafrost on flat-mounds peat and forest, with T _{average} of -0.1 to -0.5°C*	Larch, fir and birch forests on sand podzol soils	2 to 4 m

*This site exhibit two-layer permafrost structure: the upper layer of 4 to 40 m and the 2nd layer represented by relict permafrost deeper than 100 m

Table ESM-1, continued.

Site	soil	% of lakes	lithology
<u>Gyda No 1</u>	Arctic tundra soil with thin peat horizon	8 to 10 %	Marine accumulation plain: marine, lagune and lake alluvium deposits; broad alluvial river valleys
<u>Pangody/ Novyi Urengoy No 2, 3</u>	Alluvium peat gley soil in river valleys; podzols on sands and gley soils Peat gelic soils	40% of lakes in forest-tundra zone; overall range from < 10% to 60-80%	Marine, glacial, and glacial till of Salekhard formation. Complex of lake deposits of Yalbinskaya suite and Zyryano-Kurganskaya stratas along the river valley
<u>Nojabrsk/ Khanymev</u>	Podzol of northern taiga and gley-podzol soils. Peat soils on bogs	40 to 45%	Middle- Pleistocene sands and clays and upper Pleistocene – Holocene lake deposits (clays, peats).

Bibliography to Table ESM-1 :

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- Peregon, A.S., Maksyutov, N.P., Kosykh, N.P., Mironycheva-Tokareva, N.P.: Map-based inventory of wetland biomass and net primary production in western Siberia. J. Geophys. Res., 113, G01007, doi: 10.1029/2007JG000441, 2008.
- Peregon, A., Maksyutov, S., Yamagata, Y.: An image-based inventory of the spatial structure of West Siberian wetlands. Environ. Res. Lett. 4, 045014, 2009.
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- Trofimov, V.T., Bady, Yu.B., Kashperuk, P.I.: On the temperature of permafrost rock of the Gydansky Peninsula. In: Natural conditions of western Siberia, Issue no 7, Moscow State University Publ. House, p. 102-111, 1982. (in Russian)
- Sergeev, E.M., Editor : Engineering Geology of the USSR, vol. 2, western Siberia, Moscow University Publ. House, 495 pp, 1976. (in Russian)
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- Water Resources and Water Balance of the Soviet Union Territory. Gidrometeoizdat, Leningrad, 200 pp, 1967.
- Botch, M.S.: Bogs of the Tundra Zone of Western Siberia (Principles of Typology) / Types of bogs of the USSR and principles of their classification. Nauka, Leningrad, p. 146-154, 1974.

P5335 L25: Additional text is required here to complete this sentence. We revised this sentence and added more references as following: “The use of remote sensing techniques and the examination of satellite images suggest that, currently in the western Siberia, the dominant processes are the increasing number of small thaw lakes in the north and draining of large

lakes to the river network in the south of the cryolithozone (Kirpotin et al., 2009; Bryskina and Kirpotin, 2012; Polishchuk et al., 2014).”

P5336 L5: Consider revising the term “permafrost lakes”. Strictly speaking the term does not make sense. It would be more appropriate to state that the study was aimed at “extending lake sampling across different permafrost zones, or a permafrost gradient to...” We agree with the first proposition and amended the text accordingly.

P5336 L10: It would be useful here to frame this work by stating the main hypotheses. This would help the reader understand how the study design tests ideas on the factors controlling lake chemistry. The hypotheses would help the reader understand what the Authors’ believe the drivers of variation in lake chemistry to be at the front end of the paper. This is important point. We revised as following the end of Introduction:

“Based on the recently collected and available literature data, we consider the chemical composition of thermokarst lakes and ponds along a 900-km gradient of climate, vegetation and permafrost coverage in order to answer the following questions:

1) Does the variation of lake water chemical composition as a function of lake surface area in continuous permafrost zone follow the trends established earlier in discontinuous and sporadic zone?

2) Is there a latitude gradient of DOC and TE concentrations in thermokarst lakes that have the similar size (subsidence, ponds, large lakes and drained lakes)?

3) 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? “

P5337 15-20: Editorial modification – “Water objects” should be “Water bodies” – Corrected throughout the manuscript.

Some editorial attention is required on the section describing the nature and mechanisms of ground ice melt. - Last sentence of the paragraph also requires editorial attention. Are the Authors referring to thermokarst lakes in the region? If so the text may read “The largest thermokarst lakes that are located within the peat bog(s) are km-size (does this mean lakes are a km in diameter?) – Yes, and we revised the text as following: The largest thermokarst lakes of the region that are located within the peat bogs are up to several km in diameter...”

Are all the lakes underlain by talik? The majority of studied thermokarst lakes have no continuous talik under lake bottom, unlike the lakes of Alaskan boreal forest (i.e., Roach et al., 2011), or the large Siberian rivers. The exceptions are the lakes of the Gyda Peninsula that can be affected by sea influence and thus have partial connection with the underground water reservoir. We added necessary explanations in the revised text.

P5338 – A description of each stage of lake development would be very useful here since this scheme was used to stratify the dataset. We agree and added the following:

“The separation of these different stages was based on the empirical relative chronosequence of lake formation and cyclic development. Small permafrost subsidences, ponds, lakes and drainage basins investigated in this study represent the typical sequence of thermokarst thawing and lake formation in the north of Western Siberia as described previously (Kirpotin et al., 2003, 2007, 2008, 2009a, b, 2011; Pokrovsky et al., 2011, 2013, 2014, Audry et al., 2011; Shirokova et

al., 2009, 2013). The appearance of the crack in the lichen cover of the surface of the frozen mound decreases the albedo of the surface and enhances the peat degradation forming a shallow depression less than a few meters in diameter. The palsa depression is then filled by the water from the soil ice thawing. The size of the depression increases forming a shallow round pond (< 10 m diameter) which grows further into small (< 100 m diameter) shallow (< 1 m depth) lake characterized by intensive peat abrasion at the border. With further increase of the lake diameter (> 100 m), the lake border is stabilized, and water becomes less acidic and less organic. The final stage of large, mature aquatic ecosystems consists of lake drainage into another larger water system or into the hydrological network, and a subsequent formation of the lake's dry bottom with a small water body remaining in the center of the drained lake. All studied bodies of water ranged from 10 m to several km in diameter with a similar depth of 1.0 ± 0.5 m under normal precipitation/evaporation conditions."

Some information on the timescale of thermokarst lake evolution in the region would be helpful. The age of the lakes and the temporal scale of their evolution are at present unknown but expected to be similar to those of other thermokarst lakes of the world (i.e., age of several decades to hundreds years, Grosse et al. (2013), and axial increment of expanding lakes of about 1 m yr^{-1} , Burn and Smith (1990)).

Since thermokarst lake evolution is likely a continuum, a clear justification for why lake size was used as the criteria to group lakes into a "stage of development" should be provided. It follows from previous studies in this region (Kirpotin et al., 2003, 2007, 2008, 2009a, b, 2011; Pokrovsky et al., 2011, 2013, 2014, Audry et al., 2011; Shirokova et al., 2009, 2013) that, since the permafrost degradation and lake formation occurs on highly homogeneous organo/mineral substrate in essentially forest-free zone, the lakes are isomorphic and the lake diameter (or surface area) may serve as the best surrogate for the degree of ecosystem development, i.e., water body maturation. We added necessary explanation in the revised text.

Figure 1. It would be useful to indicate the approximate location of the transition zone between continuous and discontinuous permafrost on the inset map on Figure 1, and also to include a scale bar so that size of this study area can be better appreciated by the readers. We revised this Figure accordingly.

It would recommend commenting on the size catchment area relative to lake area. What is the relationship between catchment and lake area for the study lakes? This is extremely insightful remark. Given the very flat orographic context of palsa peat bogs, we studied only the closed lake basin, i.e., those that do not have any inlet and outlet. The snow thaw water and the rainwater feeding these lakes via surface inflow from June to September simply follow the relief features. Given that typically, the lakes occupy between 40 and 60 % of the territory in studies sites, we tentatively evaluate the watershed area of the lakes as equal to that of the water surface area. This is very rough estimation, and the work on GIS-based assessment of lake catchment areas is in progress. We added requested information in the revised text.

P5340 15: A few additional words are required in this first sentence to create proper context. We rephrase this sentence as following: All studied thermokarst subarctic lakes from the continental zone are likely to be fed by the atmospheric source as follows from the low concentration of major anions.

P5340 20-25: It is useful to clarify whether the Authors' believe these are subpermafrost groundwaters contributing to the lake water, or simply that water flow is through

a deeper, mineral rich active layer. We believe that this lake is fed by subpermafrost groundwaters and we explained it in the text accordingly.

Figure 2: Spelling corrections are required for “continental, rectangles, triangles and circles”. Corrected.

Clarify why the sample population was grouped by the stage of lake development for the southern part of the study region but not for the north. The reason for this different grouping is that the sequence of lake development translated into lake size (surface area) is valid for sites 2 and 3 of the south but might not be warranted for the site 1 of the north. We added an explicatory sentence in section 3.1 of revised manuscript as requested.

Table 2 – What are the correlations between the different chemical parameters? Establishing the water quality parameters that are highly correlated may help to determine which are most important to report, and which figures and discussion may be omitted. We revised the text and presentation following this constructive remark. In particular, Fig 4 of revised version reports the correlations between most important major elements whereas less important correlations are illustrated in Fig. ESM-2. We also provided in the text and in the Figure ESM-2 caption the Pearson correlation coefficients.

P5342 – I think it is important to emphasize the nature of surficial deposits, particularly the organic characteristics in this region. What is the peat thickness in the study region? The peat thickness varies from 0.2 to 0.5 m in the north to 2 to 4 m in the south. We added this missing information in revised section 3.2. Complete comparative description of the territory including forest, vegetation and sediment inventory are given now in Table ESM-1.

P5342 – 1-5 – This section requires some clarification. I assume that the Authors’ mean that lake circumference increases linearly with thermokarst expansion, whereas area increases exponentially. Consequently a lower amount of thawed materials relative to lake water volume are contributed as a thermokarst lake grows in size. I see that this is better explained on Page 5343, but this preceding section remains difficult to decipher. We agree with this remark and added an explicatory sentence in section 3.1 as recommended.

P5342 L24 – Change “objects” to “bodies”. The correct terminology should be “water bodies”. Corrected throughout the manuscript.

P5343 – L26-30; Suggested editorial adjustment to clarify that reference to DOC variation relates to work by Vincent and Pienitz, 1996 “The difference in DOC amongst coastal lakes of varying size is comparable to the northward decrease in DOC across the treeline tundra transition reported by Vincent and Pienitz (1996).” Corrected accordingly.

P5344 – L10-18: This section requires editorial attention to improve clarity of ideas. Following the recommendation, we greatly revised this part of the text and added necessary explicatory sentences.

Figure 7. Caption requires attention. Element concentration is plotted against lake diameter. We corrected the caption of this figure and extended the description of this plot (now Fig. 5 in revised version).

P5345 L4-20; P5346 L16-18 Line 16-18 indicates that “other” chemical elements are not statistically different across the different stages of lake development. This implies that some of, or all of the parameters discussed earlier are significantly different across the different stages of lake development. It should be clear which differences are significant.

In response to this comment, we added necessary statistical treatment for the difference in element concentration among different lake stage (lake surface area). The revised text in section 3.3 was amended with the following paragraph:

“This classification is based on non-parametric H-test of Kraskal-Wallis which determines whether three or more independent samples belong to the same population based on medians. In this regard, it is analogous to the parametric one-way analysis of variance (ANOVA) and in our case it shows the difference between different stages of lake development based on the concentration of each given element. It follows that for statistically significant differences are observed for 10 most abundant elements: Fe (H=12.63, p=0.0132), Na (H=19.75, p=0.0006), Mg (H=10.85, p=0.0283), K (H=24.85, p=0.0001), Zn (H=10.19, p=0.0374), and Cr (H=10.28, p=0.0360). At the same time, Si, Ca, Al and Mn do not exhibit statistically significant differences among different stages of lake development.”

To assess statistically different element ratio (Fig. 9 of 1st version, now Fig. ESM-1A), we used Mann-Whitney U Test which allows to estimate the difference between two independent set of data based on one given parameter. In our case of small number of data set, it allows to judge the difference of each element concentration between the 1st and the other stages at the significance criterion as of $p < 0.05$. In Fig. ESM-1A, the elements that met this criterion are labelled by asterisk.

Table 3. Which of these correlations are significant? Significant correlations of DOC, Fe, Si, Al, Mn, Co, Zr, Yb and Th are shown in bold in the revised version.

P5347 L22-24: When I inspect the supporting figures and the summary table I am not convinced that Rb, Sr, B and U all increase with lake diameter to the 4th stage as stated in the text. Is there a statistical basis for making such a statement?

Please see our response to your comment P5345 above. We removed B and Sr plots from revised manuscript and we rephrased the text as following: “Based on both non-parametric H-test of Kraskal-Wallis and the parametric one-way analysis of variance (ANOVA), Rb and U demonstrate statistically significant a concentration increase with lake diameter increase...”

The text throughout is a mix of results and discussion (5346-49); I cannot comment on all of the interpretation of geochemical patterns made here by the Authors, however in my view the discussion would greatly benefit from being more focused. In the revised version, the majority of discussed geochemical patterns (Sections 3.3-3.6) are supported by rigorous statistics.

P5350 L10-11: This sentence requires clarification, in particular with reference to recommendation of a world average”. We re-phrased this sentence and removed the unclear term in revised version: “Given the lack of comprehensive database on trace element concentrations in world’s lakes, we used 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) to assess the specificity of Siberian thermokarst lakes compared to other continental waters.” We also added statistical explanation for the significance of the difference between Siberian thermokarst waters and other continental waters.

P5351 L9: Please add the reference to support the statement. The most recent compilation is our book published by Nova Science Publishers (Pokrovsky et al., 2014), and we added this reference as requested.

P5352 – The Authors should provide a statistical summary of the reported patterns. From figure 12a it would be useful to understand if there are significant differences for a particular analyte, for a given lake size class across the latitudinal gradient. This point is well taken and we added a big deal of statistical treatment of results shown in Fig. 12 (now Fig. 7) of revised version. To interpret the data shown in Fig. 12 (now Fig. 7) we used Spearman's rank correlation coefficient (r_S) at $p < 0.05$ since our data are not distributed according to normal law. 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 **Figs. 7 A-L**:

- (1) A decrease in element concentration from the south to the north with a factor of 2 to 4 (DOC ($r_S = -0.39$), Pb ($r_S = -0.28$), V ($r_S = -0.27$), Ba ($r_S = -0.50$), Sb ($r_S = -0.58$));
- (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 ($r_S = -0.27$), Fe ($r_S = -0.24$), Al ($r_S = -0.54$), As ($r_S = -0.47$), REEs (La ($r_S = -0.28$), Ce ($r_S = -0.42$), Pr ($r_S = -0.43$), Nd ($r_S = -0.40$), Sm ($r_S = -0.44$), Eu ($r_S = -0.55$), Gd ($r_S = -0.49$), Dy ($r_S = -0.40$), Ho ($r_S = -0.40$), Er ($r_S = -0.38$), Tm ($r_S = -0.47$), Yb ($r_S = -0.43$));
- (3) a maximum concentration at the northern border of the discontinuous permafrost site in the continental zone of the Novyi 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 ($r_S = -0.53$), Co ($r_S = -0.56$), Cd ($r_S = -0.32$), Cs ($r_S = -0.28$), Zr ($r_S = -0.55$), Hf ($r_S = -0.55$), Th ($r_S = -0.49$); 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 ($r_S = 0.66$), Cl ($r_S = 0.52$), Ca ($r_S = 0.33$), Na ($r_S = 0.55$), Mg ($r_S = 0.43$), K ($r_S = 0.46$), Sr ($r_S = 0.43$), Cu ($r_S = 0.38$), Ni ($r_S = 0.25$) and Cr ($r_S = 0.35$)).

Figure 12. Remove “the evolution of” The figure reports “lake water pH, DOC, Ca and K concentrations...” Corrected as recommended.

P5353 L6-21: This section requires editorial attention to improve clarity. We greatly revised this paragraph in response to this comment. The latitudinal profile of DOC concentration demonstrates a general decrease [DOC], more than three times from the most southern to most northern thermokarst lakes (Fig. 7 A). This decrease is statistically significant ($r_S = -0.39$) and valid for the full range of considered lake size. Therefore, if one assumes that the increase of permafrost thawing in the north of western Siberia will provoke a shift from continuous to discontinuous/sporadic permafrost, then the concentration of DOC in small size surface water bodies may increase as much as 300%. This observation may be due to the enhanced mobilization of organic carbon from peat deposits at elevated temperatures, the increase of the duration of the active period and the increase of the thickness of unfrozen peat layer deposits feeding the lakes within the concept of soil warming scenario.

P5355 L15 Please clarify the meaning of “watershed divide of the discontinuous permafrost zone”. We rephrased this sentence as following: “Compared with lakes of site 2 (N Urengoy) and 3 (Pangody) of the discontinuous permafrost zone that are located at the watershed divide, the coastal thermokarst lakes of site 3 (Gyda) from continuous permafrost exhibit a factor of 2 to 10 lower concentration...”

P5355 L25: Clarify the meaning of this sentence. “based on the current state of permafrost rocks in the north of western Siberia:..” We rephrased this as following: “Based on the current tendency of permafrost thawing in the north of western Siberia...”

I caution the Authors about being too speculative in their Conclusions. We revised and moderated our conclusions, especially with regard to climate change consequences.