Reviewer No 1

1. *The first major comment of this reviewer is that "our conclusions are generally overdraw as they are based on a set of spatially distributed measurements of hydrochemical data of two different regions from one or two years."* This comment is well taken; we greatly moderated our conclusions and shortened by 24 lines our discussion on the consequences for climate change; we also removed the climate change context from the title and from the abstract.

Secondly, a large part of the interpretation and conclusion uses the conceptual idea of lake stages where "lake diameter" is used as a surrogate for "lake stages". However, these process studies of lake formation and succession are not provided in this paper. We do postulate that the lake diameter can be used as a surrogate for the stage of lake development and thus, in certain sense, of the lake age. This assumption is based on results of our studies in the north of western Siberia (Kirpotin et al., 2008, 2009, 2011; Shirokova et al., 2009, 2013; Aydry et al., 2011; Pokrovsky et al., 2011, 2013). The separation of these different stages was based on the empirical 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. 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)).We provided necessary explanation in revised text and properly cited the relevant publications.

The reviewer further suggested that direct parameter of "lake age" could have been dating the age of lake sediment or lake water. This is good point; however, we were unsuccessfully trying to date the lakes formation over last several years. In fact, the studied lakes exhibit very low phytoplankton production and the majority of DOC is of soil (allochthonous) origin (Shirokova et al., 2013). Both lake water and organic-rich bottom sediments are essentially represented by allochthonous organic carbon of several thousand years radiocarbon age from adjacent peat soils. To date the lake formation and evolution, therefore, modern high resolution ^{10}Be, ^{26}Al or ^{210}Pb techniques are necessary. This work is in progress but it is beyond the scope of the present manuscript.

The reviewer recommended that we "can draw results and conclusions from hydrochemical differences of lakes of two regions (North/South) and differences within these regions". This is certainly true and we did our best to orient our discussion towards the measured difference between the Gyda site and the Pangody site of this study. However, in order to put the results in the perspective of available data on thermokarst lakes from western Siberia, we also analyze the chemical composition of most southern thermokarst lakes, from Nojabrsk region (Shirokova et al., 2013). As stated by reviewer, we do discuss the differences within the regions as we plot the element concentration as a lake diameter (now lake surface area) for several stages of the Pangody site, the Gyda site (this study), Novyi Urengoy site (Pokrovsky et al., 2011) and most southern Nojabrsk site (Shirokova et al., 2013), see for instance Fig. 5 (now Fig. 3) for DOC. The discussion of the element concentration evolution is also given in the text, and we underline the similarities in hydrochemistry change as a function of the lake surface area (lake stage development) for various sites.

2. The second major comment of this reviewer is on the physical parameters used in data presentation. The reviewer is wondering, "what is the rationale to use lake diameter as physical parameter in the statistical analysis together with lake chemistry data? The hydrochemistry of the lake water is a complex interplay of the lake catchment, processes occurring in the lake and the water budget of the lake (precipitation, evapotranspiration, runoff, groundwater). As such, the reviewer recommends performing the statistical analysis using the lake volume or lake surface area. This is valuable comment and we directly followed this recommendation in revised version of the manuscript via re-drawing Figs 2, 3, 4, 5, 7 and 8 as a function of lake surface area and revising pertinent statistical treatment. The depth of the lakes is almost independent on the lake size; this is important peculiarity of western Siberia thermokarst lakes (Pokrovsky et al., 2014). As such, the results of statistical treatment remain the same after re-analysing the concentration data as a function of surface area instead of the lake diameter.

According to this reviewer, "The paper should also provide much more detail on the difference between regions 1-3 in terms of permafrost distribution, geologic material, topography, climate and hydrology (precipitation, evapotranspiration and runoff)." He/she further requested presenting additional specific properties of the lakes as for example the degree of their connection to the river. We added all necessary information in the revised version of the paper in the form of comparative Table (Table ESM-1).
It is known that connectivity between lakes and rivers is an important factor of lake area dynamics and temporal evolution in the permafrost zone (cf., Chen et al., 2013). All the lakes sampled in the late summer in this study were closed-basin lakes thus presenting the lowest seasonal variability as also follows from other studies of western Siberia (Karlsson et al., 2012) and Alaska (Chen et al., 2013). Finally, the majority of studied thermokarst lakes have no taliks under lake bottom, unlike the lakes of Alaskan boreal forest (i.e., Roach et al., 2011). 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 rectified the text accordingly.

The third major comments of this reviewer is about the latitudinal gradient. According to him/her, since the paper uses data from three study regions, two in the south and one in the north, the conclusions should not be drawn about latitudinal gradients, but about two regions, as well as differences within these regions. We basically agree. However, we do consider larger than Pangody(Novyi Urengoy) – Gyda gradient, since we analyzing the data collected from the discontinuous/sporadic permafrost zone of Nojabrsk, some 500 km south of Novyi Urengoy. Care of such a large N – S distance, we can provide some first-order conclusions on the latitudal (permafrost) gradient. Concerning the analysis of the differences between the lakes within each region, here a number of figures (Fig. 2 to 8 and Fig. 10) and pertinent discussion address the variation of chemical composition at various stage of the lake development within the site of Pangody/Novyi Urengoy. The most northern site (Gyda) does not exhibit clear division on lake stages and this site is always presented and discussed separately.

By including results of both regions in one data plot (often logarithmic plots), much of the interesting information (for example differences within the region) is lost. The research question needs to be rephrased, and accordingly, the data set should be separately analyzed and presented in figures (North/South). We re-formulated research questions as following: 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? and 2) Is there a latitude gradient of DOC and TE concentrations in thermokarst lakes that have the similar size (subsidences, ponds, large lakes and drained lakes)? The majority of collected data are from Pangody/Novyi Urengoy region and they, indeed, discussed separately from data of the Gyda Peninsula. The difference between the lakes of different size within the regions is clearly seen in Figs 2 to 6. We do not think it is worth presenting southern and northern lakes separately as it will greatly increase the total number of figures.

Finally, the reviewer noted that there are too many figures included in this paper. We removed 14 figures and moved another 7 figures from the main text to the Electronic Supporting Information.

The reviewer also suggested reformulating the major conclusions with reference to lake formation and stage hypothesis and latitudinal gradient. These conclusions are based on now well described 900-km latitudal profile presenting contrasting climate and permafrost coverage. Thanks to addition of a great deal of information on region description, this discussion is directly based on available data and contains minimum speculation. *We do agree to remove the discussion on the climate change effects. As also recommended by reviewer, in the revised version we used the lake surface area instead of lake diameter for data presentation and discussion.* We also added necessary explanations on the hydrochemical differences between the regions and within these regions and provided missing physic-geographical data (Table ESM-1).

<u>*Corrections of detailed remarks of reviewer No 1*</u>

Page 5334, Abstract Line 1-3: Remove link to thaw lake and pond formation and succession-data on this are not shown in this paper. Agree and corrected the text accordingly.

Line 9: Remove link to "lake development" - it is not discussed in this paper. Agree and corrected.

Line 12: Remove link to "dynamic succession"- it is not discussed in this paper. Replaced by 'evolution" since the conceptual scheme of these processes is well established in previous works of our group, now properly described and cited in the text.

Page 5336 Line 17: What do you mean with "surface peat dissolution"? Please clarify. Replaced by "element leaching from the peat in surface (unfrozen) soil horizons."

Study site and methods. Generally: there should be more information given on: depth and type of permafrost, parent geologic material, climate, water balance, etc. This information is now presented in section “Study sites and methods”.

Page 5337 Line 3: Simply state your first hypothesis as: Is there a latitude gradient: : :.in thermokarst lakes (remove the link to the “stage of development”). We completely reformulated these two research questions, see our response to the last major comment of this reviewer.
Line 25: Remove the linkage to lake stages and used lake diameter as the physical parameter. We partially corrected this sentence. The separation of these different stages was based on the empirical relative chronosequence of lake formation and cyclic development as now described on 20 lines of the text that follows.

Page 5338. Water samples were collected during different years (2010/2011) and potentially different seasons by including the study of Shirokova et al. 2013. Please provide more details on the interannual and seasonal variability of the hydrochemistry of the thermokarst lakes, as well as the study by Shirokova (which years and season were the samples collected?). The previous study (Shirokova et al., 2013) is based on samples collected during exactly the same period, August 2010, in the south of discontinuous permafrost zone. We added this information in revised text.

Furthermore, how is the hydrogeochemistry affected by different water balance years? This is a good point. It is known that connectivity between lakes and rivers is an important factor of lake area dynamics and temporal evolution in the permafrost zone (cf., Chen et al., 2013). All the lakes sampled in the late summer in this study were closed-basin lakes thus presenting the lowest seasonal variability as also follows from other studies of western Siberia (Karlsson et al., 2012) and Alaska (Chen et al., 2013). We alerted the reader about this possibility.
Are the years 2010 to 2011 representative for the long term mean water budget? Yes, these years were quite representative for the long-term precipitation regime and correspond to the mean water budget (see discussion in Pokrovsky et al., 2013). We added this information in the revised text.

Page 5339, Line12: Relative difference of < 30 % for B and P seems rather high; is this affecting the results and discussion? This is correct remark; however, we do not present the data on B and P in Table 2, nor in the figures. We removed this sentence from the revised text.

Results, Page 5341
Line 12: Remove “variation of different stages of lake development..” Replaced with “lake surface area”.
Line 20: Why should there be sub permafrost groundwater feeding into lakes in this continuous permafrost area? The studied lakes and ponds by Abnizova et al. 2012 have higher conductivities and are not connected to sub permafrost groundwater. The hypothesis of groundwater inflow is rather speculative, if you do not have data that indicate this (chemical, hydrogeologic). What other potential reasons could explain this interesting finding at your site? The reviewer made a good point about the analogy with lakes and ponds of the Samoilov Island (Abnizova et al., 2012). We revised our explanation of such high conductivity values. Given the proximity of the Gyda site to the sea, atmospheric aerosol deposits and local mineral substrate subjected to the influence of sea salts as it is also known

in the European Russian Arctic, could be likely explanation. We modified the text accordingly.

Tables and figures. Tables 1, 2. Instead of "stages of evolution" and "arctic lake", the lakes should be categorized by their volume or area. Following this recommendation, we added the column of lake area in this table. However, we would like to keep the term "lake stage" as this is based on existing knowledge of the lake status, now described in the section "Study site and methods" of the revised manuscript, along with pertinent references.

Figure captions. Please check the spelling of all figure captions, there are many typos (especially figure caption 2). Also, the figure captions need to completely describe the data sets, i.e., should describe the figures fully. We revised and extended the figure caption description.
Overall, the number of figures needs to be reduced. Following this remark, we removed 14 plots from the revised version and placed 7 plots from the main manuscript body to the Electronic Supporting Information.

Figure 1. The figure legend and caption are incomplete- where is study region Nr. 1? Site No 1 (Gyda) is shown by green/grey color and poorly visible in black and white mode. We explained the location in revised figure caption.
What do the grey dots (Dudinka etc) represent? Grey dots represent the towns, as now explained in the revised version. We also provided geographical coordinates, permafrost boundary, km scale, and removed the hatched backgrounds.
The reviewer correctly pointed out that "These figures also demonstrate that detailed sampling was carried out in two major regions (North/South), rather than across a latitudinal gradient." For discussion of latitudal gradient in the text, we also used results of extensive sampling in discontinuous/sporadic permafrost from Nojabrsk region, some 500 km south of Novyi Urengoy

Figures 3-8. Please provide more information on the comparative studies that you include in your graphs (Abnizova, Shirokova, Pokrovsky, Boushard). The studies are described in the text. As requested we added geographical location of other studies to revised Figures caption and we explained that results of other studies represent a powerful regression of concentration as a function of lake surface area. Showing literature data as symbols or fields of data will greatly complicate these figures making them hardly readable. Moreover, the essential message we deliver from these results is the evolution of chemical composition of the thermokarst waters with the lake size (now surface area, as requested). For this, trend lines summarizing the published data are the best way of presentation.

Furthermore, some figures include the above mentioned comparative studies; others do not (for example Figure 7a). Please be consistent. The reason of not presenting in Fig. 7a (now Fig. 5) of other literature data is the lack of these data in the original publication or the difficulty of mathematical approximating the available concentration as a function of lake surface area. In case of lack and high dispersity of lake water concentration data points for trace metals, the other studies could not be adequately represented in all figures of the manuscript. We added an explicatory sentence in revised version.

*Why are some of the symbols encircled (for example Figure 6A, 8A/B)?*The circle represents the data of arctic coastal lakes (Gyda site) exhibiting an order of magnitude higher conductivity at otherwise similar DOC. We explained it in the revised text. Fig. 8A/B was removed.

Figure 9. I recommend removing this figure. We agree and moved it to the ESM-1.

Figure 11. I do not understand the relevance of this figure. Why "two" types of lakes? I recommend removing it. Russian boreal subarctic zone comprises two major types of lakes: thermokarst lakes developed on permafrost terrain and lakes of glacial origin, most frequently studied in the European Arctic. The role of other type of lakes (tectonic, coastal, of the river valley) is small and subordinary to these two main types. For this reasons, comparison between the dominant lakes is, in our opinion, warranted. However, following the reviewer's remark, we removed this figure from the main text.

Figure 12 (Fig. 7 of revised version). In contrast to the previous figures, these figures use lake diameter (not stages) as physical variable. The reason for such a plot that it allows to include in the consideration not only samples of discontinuous and sporadic permafrost (Novyi Urengoy, Nojabrsk) where the stages and lake size are directly linked, but also the lakes of Gyda developed on continuous permafrost. For the latter site, the sequence of lake development is unknown and only lake size can be used for data presentation. We added necessary explanation in revised text.
The statistical information (differences between the regions) is lost by plotting the data on a latitudinal gradient figure. We used rigorous statistics described in the end of section 2
Furthermore, samples were collected not over the entire climate gradient, but detailed in two regions. This is correct. However, 4 sites plotted in Fig. 12 (Fig. 7 of revised version) include available literature data. Simultaneous analysis of ALL published data on western Siberia thermokarst lakes allows assessing of latitude/permafrost effect on lake water hydrochemistry. Thanks to such a complete analysis, we were able to address not only the differences between southern and northern site but place our finding in much larger geographical and climate context.