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***Interactive comment on* “Effect of permafrost thawing on the organic carbon and trace element colloidal speciation and microbial activity in thermokarst lakes of Western Siberia” by O. S. Pokrovsky et al.**

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Reviewer No 1 (Johan Ingri) The reviewer is confused, how our statement that culturable heterotrophic bacteria decrease along the lake development is compatible with the statement that there is a progressive decrease of DOC and metals. In this and previous work (Shirokova et al., 2009) we do observe a systematic decrease of both culturable bacteria number and [DOC] from the beginning of lake formation to mature (old) stages. Three explanations of this can be suggested: 1) bacterial cell accumulate in the sediments via direct precipitation or via grazing by zooplankton at the late stages

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of lake development; 2) In addition to bacterial consumption of DOC, there is a photo-induced degradation as it was also suggested by all three reviewers . Note that the shallow depths of studied lakes (0.5 to 1.5 m) favor both of these processes. Finally, as it was correctly pointed out by reviewer # 3, culturable heterotrophic bacteria may only represent a small fraction of total active bacteria in such systems. As such, the small number of bacteria we measure at the final stage of lake development does not necessarily quantify very active microbiological processes of DOC consumption occurring in the system. In response to this comment and comments of other reviewers, we added a big deal of discussion on the importance of these processes and we corrected the sentence of conclusion as following: “We hypothesize, that, upon progressive consumption of OM by different types of bacteria and photo degradation in the course of lake development, there is a systematic decrease of both DOC and dissolved metal concentrations.” This and other reviewers note that our statement that phytoplankton produce small ligands that complex Cu and Cd should be better supported, notably in view of the possibility of photo reduction processes. In the original manuscript, we hypothesize the small ligands production by phytoplankton based on available literature data on strong link between such ligands and Cu and Cd in other freshwater settings. We stated this hypothesis in the revised version of the manuscript. We fully agree with the importance of photo reduction in studied type of waters. Following this and other reviewer comments, we added a pertinent discussion with necessary references in the revised version of the manuscript. The reviewer asking us explaining the reason why the total dissolved concentrations of Fe and Al are higher than other major elements like Ca, Mg and which type of weathering or other processes can form this type of enrichment of Al and Fe. First we would like to clarify that in the text we stated that “typical concentrations of Al and Fe range from 0.1 to 1 mg/L which is even higher than the concentration of other major elements such as Mg, K, Si”. We do not consider enrichment of water in these metals compared to that of Ca, which still remains the major inorganic component. Concerning the specificity of chemical weathering in the region, we believe that the observed composition of the lake water stem from the weathering

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of already significantly altered Neogene sands underlying podzols and acid peat soils quite poor in Na, Ca and Mg and enriched in Al and Fe. Additional factors of enhanced mobility for Fe and Al in studied waters are extremely low pH rendering Al and Fe in partially ionic form and high concentration of DOM stabilizing Fe and Al colloids at elevated pH. We added a necessary explanation in the revised version (section 3.1). The reviewer further suggests discussing the ratios Fe/Al, Al/Ti, Zr/Ti between local bedrock and the lake water. This is important remark and we added a paragraph on this issue in the revised version of the manuscript: “This high mobility of usually “immobile” elements is further illustrated by comparison of dissolved and colloidal element concentration ratios (Fe/Al, 3 to 10; Ca/Al, 2 to 7; Ti/Al, 0.01 to 0.03, and Zr/Al, 0.001 to 0.003) to those measured in the soil profile of unfrozen peat (Fe/Al, 1.5-3.5; Ca/Al, 0.5 to 1.5; Ti/Al, 0.05 to 0.07, and Zr/Al, 0.002 to 0.003). The similarity of these ratios demonstrated relatively low fractionation of chemical elements between the surface fluids and the source material. Note that unlike rivers draining the mineral bedrock, the lake and ponds water located within the frozen peat terrain are unlikely to bear a signature of mineral horizons, being completely dominated by dissolution of peat organic material.” The reviewer inquires on the presence of reduced interface in the bottom sediment/solid organic soil. This is very important question. The presence of anoxic bottom layer could not be evidenced within the resolution of our measurements by submersible oxygen sensor (± 10 cm). However, our unpublished data on sediments and their pore waters composition for three lakes of this study (U-1, U-11 and U-13) (Audry et al., to be submitted) suggest the presence of a reduced interface at the first 0.5-1.0 cm of the lake bottom sediments. We added a pertinent sentence in section 3.1. The reviewer asks why are high Mn concentrations found in most lakes and whether the decrease of Mn in Fig. 9, along the chronosequence of lake development, is correlated with temperature and pH. There is no correlation between Mn concentration in studied lakes and water temperature; however, there is some weak correlation with pH. We think that the main reason for Mn decrease in the course of lake development is its link with DOM. Indeed, concentrations of divalent transition metals decrease during

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filtration and dialysis in parallel with that of DOC as illustrated for Mn in the Electronic Supplementary Information 4 (Fig. ESM-4C). In fact, more than 90% of Mn^{2+} is in the form of 1 kDa – 0.45 μm organic colloids. This may explain progressive decrease of Mn concentration along the sequence of lake development accompanied by a ten times decrease of [DOC] (Fig. 4). We added necessary explanation in section 3.3 of revised manuscript.

Technical corrections: According to reviewer, the map in Fig. 1 is not very clear and the detailed map is not necessary. We do not completely agree with this remark. The specificity of this study is that we dealing with rather small (1-10-100 m^2) size water bodies distributed over several thousand km^2 wetland which does not have large rivers, cities, roads etc. It is almost impossible to simplify the presentation of sampling points. We think that detailed map of the region is still useful because i) it gives an overall view of lake shapes and size; ii) it provides the idea of lake and pond localization with respect to bogs, forest and local topography and iii) it positions the sampled lakes with respect to local hydrological network. Note that several aerial photos of lakes are given in the Electronic Supporting Material.

We corrected the term “neutral molecules” and we shifted the elements such as Sb and As to the group of anions. Note that Ge is likely to be in the form of $Ge(OH)_4^\ominus$. The reviewer is right that in Fig. 12, the correlation coefficient is larger for Fe compared with Al and we modified the text accordingly. The reviewer is confused that in our equation for K_d between colloids and dissolved fraction, TE colloidal represent not only incorporated but also surface-adsorbed fraction. This is absolutely right and we corrected the text accordingly. The “dissolved (< 1 kDa)” is a conventionally dissolved fraction as it is always defined in the literature.

We corrected all small typos and we re-wrote the unclear sentences as recommended. We thank Johan Ingri for his insightful comments.

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