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Evolution of Fe oxides crystallinity in permafrost deposits from mid-Pleistocene to Holocene: implications for mineral organic carbon interactions

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Mineral-organic carbon (OC) interactions are involved in the geochemical stability of OC and thus in the susceptibility of permafrost to release greenhouse gases. Those so-called stabilizing or protecting interactions - accounting for ~30 % to ~80 % of permafrost soil OC - includes organo-mineral associations, such as OC sorbed onto Fe-oxides, or organo-metallic complexes. Over timescales of soil development (millennia), the capacity of soils to stabilize OC is linked to soil development through changes in soil mineralogy. Specifically, weathering products such as short-range order minerals (e.g., poorly crystalline Fe-oxides) have an extensive surface area to bind OC. However, over time, these minerals evolve towards more crystalline phases with a lower surface area available to bind OC. Freezing conditions are considered to minimize changes in Fe oxides crystallinity at short time scale, but can we consider the mineralogy of Fe oxides in a frozen deposit as stable over millennial timescale? We investigate this question along a sequence of permafrost deposits from the headwall of the Batagay megaslump, Siberia, comprising sediment up to ~650 ka old. We analyzed the proportion of Fe as poorly crystalline and crystalline Fe oxides and organo-metallic complexes, and the proportion of total OC pool forming mineral-OC interactions in the different stratigraphic units. Our data show that: (i) the proportion of iron as poorly crystalline iron oxides significantly drops with increasing age of the deposit, from 28 ± 14% for Holocene deposits to 6 ± 2% for mid-Pleistocene deposits; (ii) the proportion of iron as crystalline oxides increases from 15 ± 20% to 34 ± 2% for the same deposits, respectively; (iii) the proportion mineral-bound OC relative to the total decreases over time from 45 ± 13% to 32 ± 6 %. These findings highlight that the mineral surfaces available for OC stabilization can evolve over millennial timescales in a frozen deposit. This raises the need to better constrain mineral OC interactions for older OC exposed by abrupt thawing of permafrost such as in megaslumps.