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The next generation of climate model should account for the evolution of mineral-organic interactions with permafrost thaw

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The Earth's high latitude regions are warming twice as fast as the global average which enhances the thawing of permafrost, i.e. the perennially frozen ground which underlies about 25% of the exposed land surface in the Northern Hemisphere (Brown *et al* 1998). Permafrost thaw exposes previously frozen organic carbon (OC) to microbial decomposition with subsequent emission of the greenhouse gases carbon dioxide (CO₂) and methane (CH₄) into the atmosphere, creating positive feedback on global warming, i.e. the permafrost carbon feedback (Schuur *et al* 2015). Permafrost contains 1460–1600 GtC, almost twice the C in the atmosphere (IPCC 2019), and 15 ± 3% of that OC stock could be emitted as greenhouse gases by 2100 (Schuur *et al* 2015). On an annual basis, this would represent 22%–31% of the current anthropogenic C emissions (considering anthropogenic C emissions from IPCC 2019). Most of that C would be emitted as CO₂ but it is estimated that 2.3% of the permafrost C emissions will be released as CH₄ (Schuur *et al* 2015). This has implications for the Earth's radiative balance, given the larger global warming potential of CH₄ at the century time scale. Therefore, evidence is mounting that permafrost thaw represents a tipping element in the Earth climate system (Lenton *et al* 2019). Yet uncertainties are associated with how this will unfold.

Estimates of future C emissions from permafrost are based on simulated volumes of OC exposed upon gradual deepening of the seasonally unfrozen layer (active layer) during the growing season (e.g. Dobricic and Pozzoli 2019 and references therein). The permafrost scientific community warns that additional processes are at play and should be considered to improve estimates of future permafrost C emissions: (i) The winter season is a source of C emissions from the permafrost, and this may offset the growing season carbon uptake (Natali *et al* 2019). (ii) Soil OC loss by lateral hydrologic export indicates that C emissions not only occur from soils thawing gradually, but may occur along waterborne pathways of OC transport

in the landscape (Vonk *et al* 2019), and this may be amplified with increasing hydrological connectivity of thawing permafrost environments. (iii) About 20% of the permafrost region is sensitive to the abrupt thaw of ice-rich permafrost creating soil collapse and exposing deep OC: this not only amplifies C emissions but also increases CH₄ emissions due to the formation of lakes and wetland upon collapse (Turetsky *et al* 2020). (iv) Rapidly eroding coastlines of the Arctic ocean lead to the collapse of permafrost and frozen OC into the ocean, creating an additional pathway for C emissions (Tanski *et al* 2019). None of these four processes is currently accounted for in existing models capable of simulating permafrost C emissions.

One additional unaccounted process in the estimates of future C emissions from the permafrost is that most existing models do not resolve the many complexities involved in OC decomposition. More precisely, current models may overestimate the response of soil OC stocks to global warming by neglecting abiotic interactions between mineral surfaces and OC (Tang and Riley 2014). In Arctic soils, old OC may be as vulnerable to soil warming as young OC, suggesting that mechanisms other than the inherent stability of soil organic matter mediate the effect of warming on soil OC decomposition (Vaughn and Torn, 2019). Soil organic matter is a continuum of progressively decomposing organic compounds (Lehmann and Kleber 2015). In addition to a portion of inherently stable OC through chemical preservation and a portion of free particulate OC which is more readily available for microbial decomposition, a portion of OC is mineral-protected. Mineral protection of OC includes physical protection (OC within soil aggregates) or physico-chemical protection (mineral-associated OC). Physically protected OC is spatially inaccessible for microorganisms within soil aggregates (occluded, involving clay minerals, Fe-Al (hydr)oxides or carbonates in aggregates). Physico-chemically protected OC can be as organo-mineral associations and/or as organo-metallic complexes.

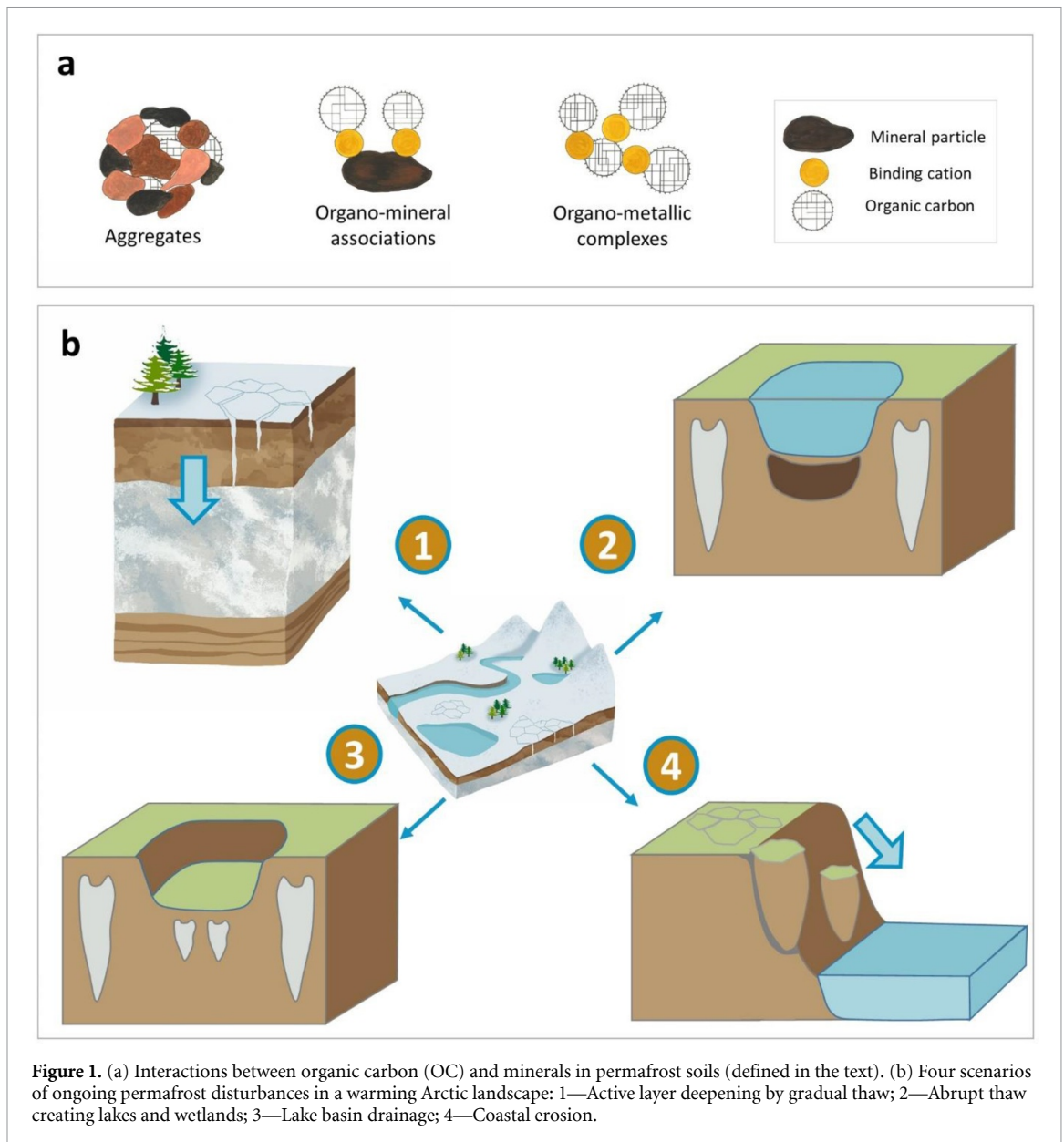


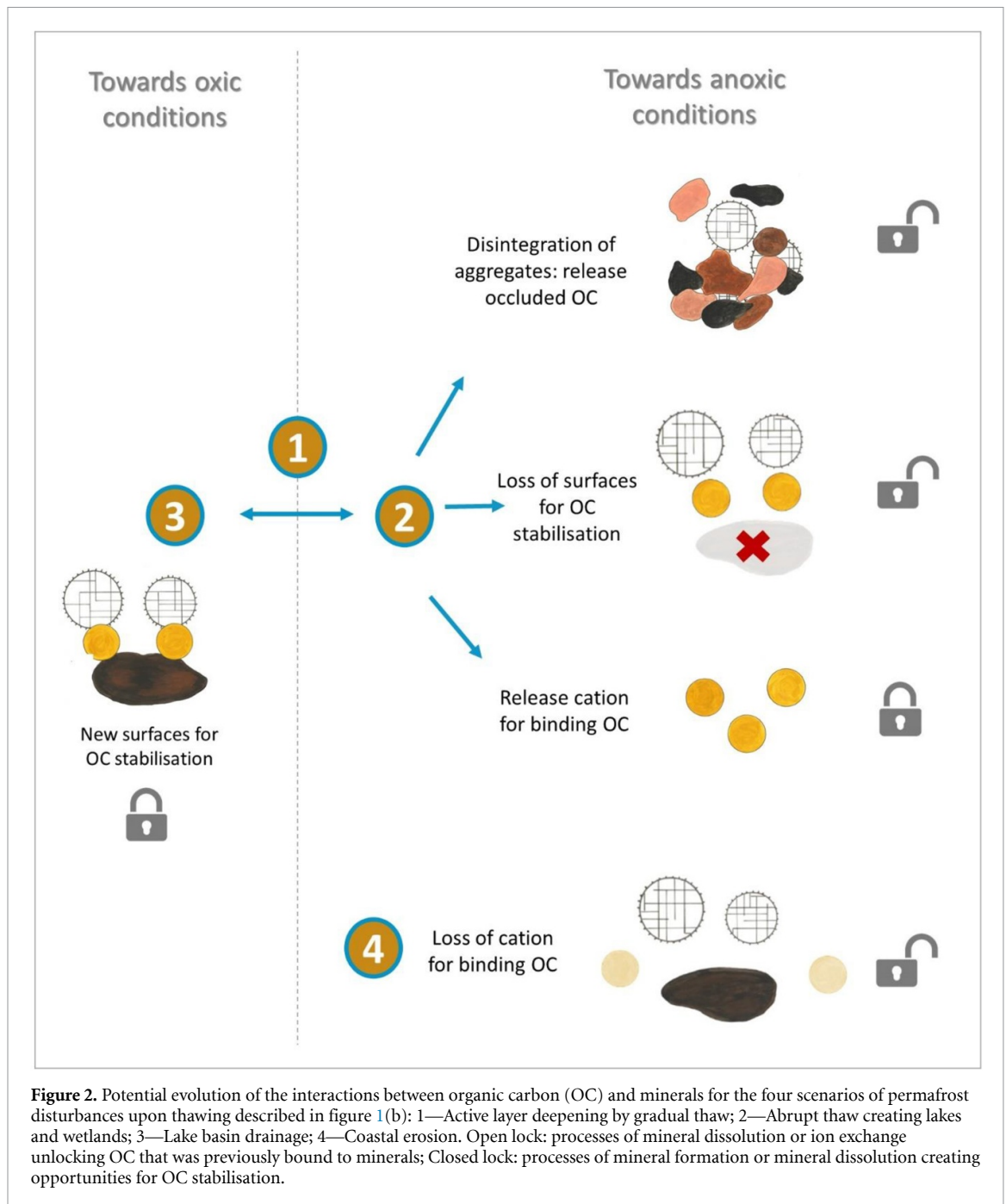
Figure 1. (a) Interactions between organic carbon (OC) and minerals in permafrost soils (defined in the text). (b) Four scenarios of ongoing permafrost disturbances in a warming Arctic landscape: 1—Active layer deepening by gradual thaw; 2—Abrupt thaw creating lakes and wetlands; 3—Lake basin drainage; 4—Coastal erosion.

Organo-mineral associations result from the interaction of OC with mineral surfaces such as OC sorbed onto clay minerals or Fe-oxides, using cation bridges such as Ca or Mg. Organo-metallic complexes result from the complexation of OC with metal ions (i.e. OC complexed with e.g. Al, Fe, Ca) (figure 1(a)).

Estimates of mineral-associated OC in permafrost soils range from ~30% (Mueller *et al* 2015) to ~80% (Dutta *et al* 2006). These interactions between OC and minerals influence the accessibility of OC for microbial decomposition and OC stability in permafrost soils (Gentsch *et al* 2018). Experimentally determined C emission rates consider that the estimated volume of OC exposed upon thawing does not necessarily correspond to the OC available for microorganisms, but do not account for the evolution of the interactions between OC and minerals upon thawing. Mineral constituents will be increasingly exposed to changing water saturation in response to

permafrost thaw and deepening of the active layer (Lawrence *et al* 2015). The mineral surfaces and metal ions binding OC are sensitive to changing physico-chemical conditions such as water saturation or soil acidity (Lehmann and Kleber 2015). These conditions are expected to vary upon thawing along the OC transport pathways in soils, along waterborne pathways, or at sea. These changing interactions should be considered to predict the evolution of OC accessibility for microorganisms and C emissions upon thawing.

Considering four scenarios of ongoing permafrost disturbances in the warming Arctic (figure 1(b)), it is evaluated how changing the physico-chemical conditions for the permafrost mineral constituents may affect interactions between mineral surfaces and OC (figure 2), thereby modulating OC availability for microorganisms, and hence C emissions from thawing permafrost and the CO₂ to CH₄ ratio of C emissions.



Active layer deepening by gradual thaw can lead to the release of soluble cations (Ca, Mg, Na) from the permafrost given that their concentration can be one to two orders of magnitude higher than in the active layer (e.g. Kokelj *et al* 2002), and these cations could serve as cation bridge in the formation of OC associations with mineral surfaces. Alternating oxic and anoxic conditions in the active layer of permafrost soils are expected with future changes in water table depth and drainage, resulting in the formation of Fe-oxides (oxidation of Fe) or dissolution of Fe-oxides (reduction of Fe) as shown in Arctic soils (Herndon *et al* 2017). The loss of Fe-oxides would contribute to the disintegration of soil aggregates releasing occluded particulate OC, and to

the loss of mineral surfaces available for association with OC. The formation of Fe-oxides would create mineral surfaces for association with OC. In parallel, more acidic conditions in wetland soils would favor the release of Fe and Al which could form complexes with OC. Overall, a portion of OC can be unlocked and transferred into a pool of freely available OC, which may contribute amplifying C emissions from permafrost to the atmosphere. Conversely, the concomitant release of metal cations upon permafrost thaw and the formation of Fe-oxides may partly mitigate permafrost C emissions by stabilisation of OC via complexation or sorption onto mineral surfaces, and thereby reducing OC availability for microorganisms.

Abrupt thaw creates lakes and wetlands, generating anoxic conditions. As in the previous scenario, the loss of Fe-oxides by Fe reduction would unlock OC that was not directly available to microbial decomposition, and would also release Fe and Al, that form complexes with OC under acidic conditions and reduce OC availability. The relative amplitude of these processes upon thawing will modulate the availability of OC for microbial decomposition and thereby the C emissions derived from abrupt thaw. Importantly, abrupt thaw increases the proportion of CH₄ emitted relative to CO₂ due to the anoxic conditions, but a portion of CH₄ emissions could be mitigated by the anoxic oxidation of methane mediated by the presence of Fe-oxides (acting as electron acceptor), as reported in Alaskan peatlands (Miller *et al* 2019). To what extent Fe-oxides modulate the CO₂ to CH₄ ratio of C emissions upon abrupt thaw is an important parameter to consider in the predictions of future permafrost C emissions.

Draining a thaw lake basin, a common phenomenon in the evolution of thaw lakes in modern and Holocene periods, leads to oxic conditions, which favors the formation of Fe-oxides. As in the first scenario, this would create mineral surfaces for association with OC, and contribute reducing the availability of OC for microbial decomposition.

Coastal erosion exposes permafrost to changing salinity, which is likely to untie cation bridges in organo-mineral associations by an exchange between e.g. Ca and Na (Blattmann *et al* 2019). This would contribute to release OC associated with mineral surfaces, increasing the availability of OC for microbial decomposition.

The analysis of these scenarios demonstrates that the stability of mineral surfaces and the availability of metal ions for binding OC are likely to vary upon thawing. The evolution of mineral-organic interactions with permafrost thaw is a potentially important player for the modulation of permafrost C emissions by affecting (i) the availability of OC for microbial decomposition, and thereby amplifying or mitigating C emissions, and/or (ii) the CO₂ to CH₄ ratio of C emissions, and hence the resulting net global warming potential.

Not considering the evolution of the interactions between minerals and OC upon thawing is likely to lead to over-confident projections of permafrost C emissions. To collectively move towards improved model parametrization to simulate permafrost C emissions, I encourage coordinated observations of OC and mineral fractions in disturbed and undisturbed permafrost regions, and the development of routine approaches to quantify changes in mineral-organic interactions with ongoing permafrost thaw. Overall, this challenge requires bridging disciplines across soil science, hydrology, geochemistry, climate modeling, cryospheric science, microbiology, and quaternary geology.

Improving models capable of simulating permafrost C emissions is part of the ongoing effort to include permafrost C emissions into the next generation of Earth system models used to predict the rate of climate change. Permafrost C emissions were not included in the Earth system models analysed by the Intergovernmental Panel on Climate Change in the last assessment report (IPCC 2013). An encouraging step is that permafrost OC stocks are now quantified in the recent Special Report on the Ocean and Cryosphere in a Changing Climate, and potential permafrost OC pool change following contrasted Representative Concentration Pathway scenarios are included (IPCC 2019).

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Data availability statement

Any data that support the findings of this study are included within the article.

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