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Speciation, mobilization, and attenuation of redox-sensitive metal(loid)s in thawing permafrost

ELLIOTT K. SKIERSZKAN[1], SARA ELLACOTT[1], MATTHEW FELLWOCK[2], DR. VALERIE A. SCHOEPFER[2], BAS VRIENS[3], DANIEL S. GRÉGOIRE[1] AND MATTHEW LINDSAY[2]

[1]Carleton University
[2]University of Saskatchewan
[3]Queen's University

Thaw of organic-rich permafrost soils drives development of anoxic conditions due to biodegradation of labile organic carbon and water-saturated conditions. Our geochemical and mineralogical investigations of permafrost cores from a remote region of western Yukon, Canada show that incipient permafrost thaw under anoxic conditions drive a rapid initial release of up to 3,200 mg L^{-1} DOC, 40 mg L^{-1} Fe, 1.9 mg L^{-1} Mn, and 96 µg L^{-1} As into porewater. These Mn and As concentrations are approximately one order of magnitude higher than Canadian drinking-water guidelines. We performed additional laboratory incubations under partially oxic, anoxic, and anoxic+sulfate-amended conditions to constrain metal(loid) mobilization under variable redox environments. These experiments also mobilized elevated Fe (up to 42 mg L^{-1}), Mn (up to 11 mg L^{-1}), and As (up to 340 µg L^{-1}), particularly under anoxic conditions. Sulfate amendments drove substantial As attenuation over time, presumably through microbially mediated As-S interactions. Laboratory column experiments using similar permafrost cores constrained temporal evolution of Fe, Mn, and As mobilization under active-flow conditions, resulting in an initial flush of these metal(loid)s along with DOC that was sustained over several pore volumes. Synchrotron X-ray absorption (XAS) and X-ray fluorescence analyses and tandem high-performance ion chromatography-inductively coupled plasma mass spectrometry indicate the presence of reactive arsenate and arsenite in permafrost soils that are liberated during Fe-(oxyhydr)oxide reductive dissolution. Ongoing XAS analyses are targeting Fe, Mn, and As geochemical evolution during incubation and column experiments, and metagenomic analyses are under way to constrain microbial drivers of Fe, Mn, and As cycling during permafrost thaw. These results collectively show highly reactive biogeochemical processes during incipient stages of permafrost thaw that mobilize nutrients and hazardous metal(loid)s, and signal a major shift in metal(loid) biogeochemical cycling in a warming cryosphere.