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Oxidation Paradox: Influx of oxidant stimulates microbial metal/radionuclide reduction

KARRIE A. WEBER[1,2], JEFFREY P WESTROP[2], POOJA YADAV[2], TAYLOR ROSSO[2], VINCENT NOËL[3], ARJEN VAN VEELEN[4], JOHN BARGAR[5], XIAOQIN WU[6], ROMY CHAKRABORTY[6] AND JOSH HERR[2]

[1]1104 T Street
[2]University of Nebraska-Lincoln
[3]SLAC National Accelerator Laboratory / Stanford University
[4]Los Alamos National Laboratory
[5]SLAC National Accelerator Laboratory
[6]Lawrence Berkeley National Laboratory
Presenting Author: kweber2@unl.edu

The existing paradigm describes oxidation of reduced chemical species, such as iron(II) and uranium(IV), following an influx of oxidants such as dissolved oxygen (DO) or nitrate. Prior field results challenged current understanding of the oxidant's role controlling redox behavior of metals/radionuclides; low concentrations of an oxidant (DO) injected into a biostimulated reduced region of an alluvial aquifer stimulated a decrease in aqueous uranium concentrations *in situ* [1]. Here we experimentally test the impact of a highly soluble oxidant, nitrate, on metal/radionuclide redox state using organic-rich, naturally reduced-uranium bearing oxbow lake sediments. Batch reactors of reduced sediments preincubated with uranyl chloride as a redox tracer were amended with and without the addition nitrate at varying concentrations (low nitrate <14 mg/L-N> high nitrate). High nitrate amended batch reactors increased dissolved uranium(VI) consistent with oxidation of reduced species. However, low nitrate amendments stimulated a decrease in dissolved uranium(VI), consistent with reduction. No significant change was observed in reactors amended with anoxic deionized water. XANES analysis of sediments supported uranium(VI) reduction with the precipitation of uranium(IV). An increase in aqueous iron(II) further supported the onset of reducing conditions. Reduction activity occurred concurrent with an increase in dissolved organic carbon (DOC) and cell and virus abundance. Batch reactors were amended with the antibiotic chloramphenicol demonstrated suppression of nitrate and uranium reduction supporting this as a microbially catalyzed process. Metagenome assembled genomes from the microbial community revealed the metabolic potential indicating complex carbon degradation, fermentation, mineralization as well as the potential for anaerobic respiration of nitrate, metal/radionuclides, and sulfate. Recovery of metagenome assembled virus genomes from microbial (>0.2mm) and virome (<0.2 mm and >0.05 mm) samples indicated a change in viral community in response to nitrate amendments and viral-encoded carbohydrate active enzymes suggesting a microbial and viral liberated carbon. Together these results indicate that an influx of an oxidant can lead to the increase in DOC and carbon cycling supporting microbial activity and reducing conditions.

[1] Pan et al., *Environ. Sci. Technol*. 2018. 52: 8133–8145.