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Effects of Fe(III) Inputs on the Rate of Methanogenesis in Wetland Sediment Microcosms

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The presence of alternate terminal electron acceptors (TEAs) such as sulfate or ferric iron has previously been shown to inhibit methane production in transiently anoxic environments. This effect has been ascribed primarily to the ability of sulfate and iron reducers to outcompete methanogens for common electron donors (e.g., acetate, H_2) because, at standard state, respiration involving these TEAs provides a greater thermodynamic driving force compared to methanogenesis. In addition, evidence exists that methanogens can transfer electrons to ferric iron and dissimilatory metal reducers such as *Geobacter* have themselves been shown to transfer electrons directly to methanogens. The perceived role of competitive inhibition in controlling methanogenesis, however, is largely predicated on experiments where the production of methane was inhibited in microcosms that had ferric iron or sulfate present at the beginning of the experiment. In dynamic redox environments like wetlands, the formation of ferric iron is instead controlled by periodic influxes of O_2 into reduced sediments, so the impact of iron oxide addition on actively metabolizing methanogenic communities remains unclear.

We conducted microcosm experiments using sediment from a freshwater wetland amended with acetate. Shortly after the onset of methanogenesis, we amended each microcosm with ferric oxide: ferrihydrite, lepidocrocite, or goethite. We observed that the rate of methane production declined dramatically immediately following the addition of iron oxide despite the continued availability of acetate. The rate of acetate consumption also decreased significantly concomitant with the addition of ferric iron. Gas measurements also showed an immediate decrease in the partial pressure of H_2 in the headspace of microcosms following the addition of ferric iron. Greater inhibition of methanogenesis was observed with the less crystalline iron minerals ferrihydrite and lepidocrocite relative to the more crystalline goethite. X-ray diffraction showed the formation of the secondary ferrous minerals siderite and magnetite in ferrihydrite- and lepidocrocite-amended microcosms, but not in those amended with goethite. Unamended microcosms were dominated by 16S rRNA gene sequences classified as *Methanomicrobia*, a class of methanogenic archaea that comprised 32% of the total community during peak methane production. In iron-amended microcosms, however, these same taxa initially accounted for roughly 10% of the total community prior to the addition of ferric minerals, but sequences classified as *Geobacter* dominated soon thereafter. These results suggest that the impact of ferric oxides on methanogenesis is more complex than just thermodynamic limitations or kinetic inhibition and requires further study.