

Effects of elevated CH₄ and CO₂ on the mobility of metals in near-surface sediments

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Carbon capture, utilization and storage (CCUS) is one practice to reduce atmospheric CO₂ emissions by capturing CO₂ from stationary, sources, transporting it to suitable storage sites, and injecting it into deep, geologic reservoirs for enhanced hydrocarbon recovery and storage. One concern with respect to CO₂ storage is that unintentional CO₂ release to the surface may occur through direct seepage pathways such as fractures and/or improperly plugged and abandoned wells.

We hypothesize that CO₂ migration into the vadose zone and subsequent Eh and pH changes in partially saturated sediments could mobilize metals and contaminate ground- and surface-waters; however, this contamination potential has not been addressed. The goals of this study are to understand if metal mobilization through soil pore water may occur due to CO₂ release and to assess the potential for impact to underlying aquifers and/or the biosphere. The study was conducted at a CCUS site in Cranfield, MS, where localized (10m²) seepage of CH₄ (45 %) from depth reaches the surface and oxidizes to CO₂ (34%) in the vadose zone near a plugged and abandoned well. Four sediment cores (4.5-9m long) were collected in a transect extending from a background site through the area of anomalous soil gas and were sampled at approximately 1-m intervals. Samples were analyzed for Eh and pH in the field and for occluded gas concentrations, metal concentrations (water-mobile, acid-mobile, and total), moisture content, and carbon content (inorganic and organic) in the lab. Data from the background area (occluded gas ~21% O₂, <1% CO₂, 0% CH₄) showed oxidized conditions (Eh from 464-508mV) and neutral pH (7.0-7.8) compared to samples collected near the soil gas anomaly (13-21% O₂, 0.1-5% CO₂, <0.1% CH₄) that were more reducing (Eh 133-566mV) and of lower pH (5.3-8.0). Strong correlations were found between Eh and O₂ (r=0.95), pH and CO₂ (r=-0.88), and between these parameters and acid-leached metals in samples from within the soil gas anomaly. Correlations quickly weaken and disappear away from the anomaly and in the background area. While total metal concentrations and the percent of water-mobile metals are relatively consistent between cores, the percent of acid-mobile metals increases toward the gas anomaly. Preliminary conclusions are that 1) Oxidation of CH₄ to CO₂ depletes O₂ causing reducing conditions; 2) high CO₂ and low O₂ alter the Eh and pH of sediments affecting metal mobility; 3) geochemical alterations are localized to the extent of the gases. Additional work will use modeling to predict the potential outcomes of altered sediment geochemistry observed in the field on aquifers – specifically to assess the potential for metals to exceed the maximum contaminant levels (MCLs) set by the EPA in underlying drinking water.

Keywords: Metals, soil, redox, carbon dioxide, methane, oxygen, pH, Eh