Integrated field, experimental, and modeling perspectives of the geochemistry of geologic carbon sequestration

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Geologic sequestration of CO₂ generated by coal-fired power plants is a critical component of Carbon Capture and Storage. Injection of supercritical CO₂ into a brine aquifer shifts rock-dominated reaction systems to fluid-dominated systems controlled by acid-generating reactions and mixed-fluid equilibria. Increased carbonic acid content in the brine reduces the pH of *in-situ* brine by approximately 1.5 to 4 pH units, depending on brine chemistry, formation lithology, and temperature. Injected supercritical CO₂ controls pH, leads to the dissolution of carbonate minerals, and perturbs the aqueous geochemistry of the system until the reservoir rock reasserts control. Small amounts of SO₂ (<1%), a common impurity in a CO₂ waste stream, alter the sulfur geochemistry of CO₂-water-rock systems. Co-injected supercritical CO₂-SO₂ further dissolves calcite and precipitates anhydrite. Natural CO₂ fields, such as the Madison Limestone of southwest Wyoming, are "natural analogues" for carbon storage reservoirs. These analogues provide the opportunity to evaluate and validate the relevance of laboratory experiments and computer models.