

Tracking CO₂ migration through a sandstone aquifer using Sr isotopes: Chimayó, New Mexico, USA

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The feasibility of storing CO₂ in geologic formations relies on our ability to understand the effects of high-CO₂ fluids on the storage formation and on impermeable seal rocks. Natural analogue sites provide an opportunity to observe effects of upwelling CO₂ on aquifer geochemistry. Wells in Chimayó, NM tap groundwater from the Upper Oligocene to Miocene Tesuque Formation of the Santa Fe Group, which is cut by faults that act as conduits for deeply sourced CO₂. Waters in the region can have elevated levels of dissolved CO₂, including a geysering well with 1.8 g/L CO₂.

Strontium isotope signatures of Chimayó well waters were used to track brine migration and quantify water-rock interactions. ⁸⁷Sr/⁸⁶Sr ratios range from 0.7176 for CO₂-charged brackish water (TDS up to 7,000 mg/L) to 0.7098 for low-TDS groundwater. Sr isotope compositions of sequentially extracted ammonium acetate and acetic acid leachates of unconsolidated and cemented Tesuque Formation lithosome samples indicate that most low-CO₂ Chimayó water chemistry can be explained by interaction with lithosome minerals. However, one high-PCO₂ well water has an ⁸⁷Sr/⁸⁶Sr of 0.7154, significantly higher than the lithosome leachates. Sr isotope mixing models indicate admixing of up to 5% of CO₂-charged brine, which could have migrated into the aquifer along the nearby Roberts Fault.

Two wells <150 m apart, located northwest of this fault, have similar ⁸⁷Sr/⁸⁶Sr ratios of 0.70979±2, significantly lower than the CO₂-brine value. However, these wells have very different CO₂ concentrations (0.3 vs. 1.3 g/L) and alkaline earth contents (0.8 vs. 45 ppm Ca). This suggests that a CO₂-resistant barrier divides the lithologically distinct unit tapped by these wells; leakage of CO₂ into one side could result in increased dissolution of carbonate cement. Strontium isotopes demonstrate that the influx of CO₂ into the aquifer in this case must have been completely decoupled from the deeper saline carrier waters observed at other sites.

Molecular simulations of the role of bridging oxygen on wet porous silica surfaces on proton transport

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Electrochemical studies have shown enhanced proton transport in mesoporous silica containing water. Achieving an accurate description of proton transport in such atomistically large and structurally complex systems requires application of dissociative potentials in molecular simulations. A dissociative water potential that matches many bulk water properties was applied to nanoconfined water, reproducing the change in thermal expansion of water confined in nanopores of silica, enabling a molecular explanation of the mechanism. The dissociative chemisorption of the water molecules on the silica surface show hydroxylation consistent with experiment and the enhancement of hydronium formation at these surfaces, similar to ab-initio molecular dynamics simulations. In addition to the formation of surface silanol (SiOH) sites, where protons are strongly bound, the simulations also show the location of weakly binding proton adsorption sites on the silica surface that contribute to enhanced proton transport beyond that observed in the nanoconfined water phase alone. These sites are on bridging oxygen (BO) with a Si-O-Si bond angle in the range similar to that obtained in molecular orbital (MO) calculations, providing an explanation of increased proton transport observed experimentally. Figure 1 shows the BO energy and the concentration of H on BO (BOH) vs Si-O-Si bond angle on silica surface exposed to water, with an energy minimum at angles similar to MO calculations at a maximum concentration of sites.

