

Tracing CO₂ and H₂S sequestration in a basaltic aquifer using stable isotopes at CarbFix, Iceland

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The last IPCC report stressed the importance to use Carbon capture and storage (CCS) to reduce the effects of anthropogenic greenhouse gases. The goal of our study is to investigate the utility of metal stable isotopes to monitor geochemical reactions induced by injection of CO₂ and H₂S in a basaltic aquifer. We have analysed Fe, Cu, Zn and Sr isotopes in water samples from the CarbFix site (Hellisheiði, Iceland) to complement existing Ca and Mg isotope data. Pilot injections of (1) 175 tons of CO₂ and (2) 73 tons CO₂+H₂S gas mixture into a deep aquifer were undertaken in 2012. Previous studies have shown that the mobilisation of cations (Ca, Mg, Fe) related to the arrival of the carbon-enriched, low-pH waters, was caused by dissolution of the formation basaltic rock. These element concentrations decreased rapidly after this initial dissolution, being trapped in carbonates, clays, zeolites and hydroxydes.

We observe substantial variations in $\delta^{56}\text{Fe}$ (>1.8 ‰) and $\delta^{66}\text{Zn}$ (>1.4 ‰) water compositions through the injections and the following monitoring period. Stable Sr, Mg and Zn do not correlate with any specific mineral saturation index, suggesting a multi-mineral influence on their isotopic compositions. Indeed, these isotopes will be fractionated by zeolite, clay, sulfide and carbonate precipitation, for which individual influence are challenging to disentangle. However, Fe and Ca isotope compositions covary when pure CO₂ is injected, along with pH variations. Co-injection with H₂S at the second injection decouples Fe and Ca isotope fractionation. Ca isotopes are shown to be controlled by calcite dissolution/precipitation at the CarbFix site. Fe isotope fractionation may then be primarily controlled by carbonate precipitation when pure CO₂ is involved while being also affected by other phases, in particular sulfides, when H₂S is co-injected with CO₂. This study demonstrates that Ca and Fe isotopes are the best candidates to quantify carbon capture in carbonates.