

Mantle CO₂ fluxes and carbonate fixation in volcanic hydrothermal systems in Iceland

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Most of the Earth's natural CO₂ degassing from the mantle to the surface occurs within the major rift zones. Fixation of CO₂ in crustal carbonates may impact the efficiency and scale of such CO₂ emissions. However, the amount of CO₂ fixed and emitted within rift zones remains mostly unconstrained. Using the Iceland rift as an analogue to the submerged mid-Atlantic ridge, we use carbon abundance and isotope systematics in hydrothermal fluids, whole rocks and carbonates from rift-related geothermal systems to unravel the origin of CO₂ and quantify crustal fixation into carbonates within rift settings.

The targeted geothermal systems included the seawater dominated system at Reykjanes (SW Iceland) and the meteoric water dominated systems at Nesjavellir (SW Iceland) and Krafla (N Iceland). Hydrothermal fluids, altered whole rock and carbonate grains were collected from the wells drilled into the

subsurface of these systems. Fluids showed large variations in the CO₂ concentration (267-3098 ppm) but insignificant δ¹³C-CO₂ differences (-4.8 to -2.5‰). In contrast, large δ¹³C variations of carbonates (-14.5 to +0.5 ‰) were observed. Carbon contents of altered rocks ranged from <0.01 to 4.33 wt.%. Thereby, carbon addition occurred within the upper 1000 m of the geothermal system whereas major losses of carbon were found at depths >1500 m.

Coupling the dataset with isotope and geochemical modeling revealed that carbon originates from a mantle source. Depressurization boiling of ascending hydrothermal fluids and water-rock interaction was found to be the main mechanism leading to carbonation. Only <1 to ~20% of the total mantle CO₂ degassing through the systems is sequestered into carbonates whereas the majority of the CO₂ is emitted to the surface through hydrothermal fluids, soil degassing and dissolved into groundwater. Active high-temperature geothermal systems in rift zones are thus inefficient in sequestering CO₂ and that surface CO₂ emissions closely resemble mantle flux and carbon isotope content fed into the systems.