Isotope geochemistry of CO_2-rich mineral springs – Natural analogs for a leaking carbon sequestration scenario

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CO_2-rich mineral springs are recognized as natural analogs for leakage from geologic carbon repositories. The geochemistry of CO_2–rich mineral springs in the western U.S. and Tibet are used to describe the origin and evolution of volatiles in these springs in order to identify isotopic tracers for use in geologic carbon sequestration. Springs often issue along normal faults and are associated with Pleistocene-Holocene travertine accumulations indicating long-term CO_2 migration. Spring waters range in pH from 5.5–8.5, from 6–58°C, are dilute to saline (100 to >20, 000 ppm TDS) and have alkalinities of 100 to >3000 mg/l (as HCO_3^-). Dissolved gases in springs are dominated by CO_2 (≤99% by volume) and/or N_2, with lesser amounts of H_2S, O_2, H_2, He, Ar, and CH_4. The $\delta^{13}C$ of CO_2 leaving springs ranges from -21 to -0.6 ‰ (vs. PDB), while the $\delta^{13}C$ of DIC in water ranges from -12.5 to +13.5 ‰. Paired measurements are necessary to evaluate the degree of CO_2 degassing and initial fluid carbon isotope composition. The $\delta^{15}N$ of dissolved N_2 gas ranges from -0.9 to +2.2 ‰ (vs. Air). Helium isotope ($^3He/^4He$) and $CO_2/^3He$ ratios from Tibet are ~0.02 R_A and ~10^{12}, respectively, indicating a crustal source. Ratios from the western U.S. ($^3He/^4He$ = 0.1-1.2 R_A; $CO_2/^3He$ = 10^9-10^{13}) indicate a range of sources from crustal to mantle. Based on these natural tracers, the sources of CO_2 are metamorphic, organic and magmatic. Using these data, estimates of carbon loss range from 100's to 1000's of kg/yr for individual springs. Regional scaled estimates for diffuse loss along fault zones to shallow aquifers and ultimately the atmosphere can grow to the order of 0.01 gigaton of C/yr. This is approximately 1% of the current annual anthropogenic addition to the atmosphere.

SIMS U-Pb dating of alluvial deposits using authigenic and detrital opal

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The use of uraniferous opal as young as 1 Ma for U-Pb dating was suggested by Ludwig *et al*. [1]. More recently, opal has been used to date a variety of low-temperature, near-surface materials. Secondary-ion mass spectrometry (SIMS) was used in this work to determine 1.5–11 Ma U-Pb isochron ages of authigenic and detrital opals from alluvial deposits in Midway Valley, southern Nevada.

Samples of authigenic opal (N=12) from pedogenic cements and clast rinds, and detrital clasts of opal (N=4), were collected from drill core of basal alluvium near the alluvium/bedrock interface 25–52 meters below the land surface. The relatively large U concentration (tens of μg/g), minimal common-Pb and Th concentrations, and negligible radiogenic $^{20\circ8}Pb$ allowed $^{206}Pb/^{20\circ8}Pb$-$^{238}U/^{20\circ8}Pb$ isochron dating by SIMS (http://shrimprg. stanford. edu). The 18–90 nA primary $^{16}O^-$ beam produced pits ~30 μm in diameter and 15–20 μm deep, which equates to ~30 nanograms of opal consumed during a 25-minute spot analysis. Measured Pb/U ratios were corrected for instrument bias by factors of 0.6–0.8, determined from U-Pb isochron slopes obtained for a 1.915-Ma opal standard analyzed during the same analytical sessions. Corrections for initial ^{234}U excess (excess ^{234}U incorporated during formation of opal results in unsupported ^{206}Pb) were applied using both measured $^{234}U/^{238}U$ and a value of 1.54 for the initial $^{234}U/^{238}U$ activity ratio of soil water determined by $^{23\circ}Th/U$ dating of young soils at the site. The age of basal alluvial deposits at Midway Valley is constrained by the youngest disequilibrium-corrected ^{206}Pb-^{238}U isochron age of detrital opal (7.29±0.64 Ma) and the oldest age of authigenic opal (3.20±0.42 Ma).

[1] Ludwig *et al*. (1980) *EPSL* **46**.