How iron and arsenic reduction pathways in metal-reducing bacteria influence arsenic fate and transport

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A global health crisis has emerged due to the consumption of drinking water derived from arsenic contaminated aquifers. Iron(III) and arsenate reducing bacteria are implicated in processes that cause arsenic mobilization in aquifers sediments enriched in arsenic. Under oxic conditions arsenate predominates and is often adsorbed onto mineral surfaces such as amorphous iron oxide. However, under anoxic conditions metal-reducing bacteria can respire both iron(III) and arsenate, generating iron(II) and arsenite. It is unclear which pathway has the greater influence on the release of arsenic. To address the problem, genetic and geochemical approaches are being used to identify which metal reduction pathway has the greater influence over arsenic fate and transport. Using a model iron and arsenate-reducer, *Shewanella* sp. strain ANA-3, strains deficient in iron or arsenic reduction were generated and tested for their abilities to alter arsenic fate and transport within batch and hydrodynamic conditions. We inoculated various *Shewanella* iron and arsenate reduction deficient strains into anaerobic bottles with slurries of ferrihydrite. Iron(II) and dissolved and adsorbed arsenate and arsenite were monitored over time. Our results showed that the arsenate reduction mutant (ARM) did not reduce solid phase arsenate even though it could still reduce iron. The iron-reduction mutant (FERM) could reduced arsenate to arsenite similarly to the wildtype strain and did not reduce iron even after extended incubation (144 hours). We also investigated arsenic transport by quantifying arsenic mobilization using flow through columns containing arsenate and iron oxide coated sands inoculated with either FERM or ARM strains. These results showed that arsenate and iron(III) reduction occur independently, and that arsenate reduction is the dominant process controlling arsenic release. Currently we are using molecular techniques to determine whether the arsenate and iron genes are differentially expressed when cultures of ANA-3 are amended with iron and arsenate.

Calcium isotope fractionation in cave-analogue conditions

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Speleothems are proving to be fundamental archives for high resolution palaeoclimate studies, but quantitative understanding of their geochemistry in relationship to climate variables is still rather poor. Laboratory and marine studies of Ca isotopes suggest that speleothem $\delta^{44/42}$Ca may act as a proxy for past cave temperature and/or the amount of prior-calcite-precipitation (PCP), but no systematic study of speleothem Ca isotope fractionation has yet been performed.

We have measured the $\delta^{44/42}$Ca of laboratory-precipitated calcite, grown in an experimental setup designed to closely replicate speleothem precipitation. Calcium solutions with a constant saturation index of 0.34 were dripped onto calcite-seeded glass plates, at tightly-controlled temperatures. There is no significant difference in the $\delta^{44/42}$Ca offset between precipitates and initial solutions at four different temperatures (7–35ºC; ANOVA, p=0.94) and three drip rates (0.18–1.22 ml/min; ANOVA, p=0.49), with a mean precipitate-solution difference of $\delta^{44/42}$Ca = -0.82 ±0.11‰. Our results provide insight into the fractionation of Ca isotopes during carbonate growth. The absence of any temperature or growth-rate influence on $\delta^{44/42}$Ca suggests that, in the natural setting, $\delta^{44/42}$Ca will be controlled predominantly by PCP. This may provide a proxy specific to this one environmental factor, and serve as a powerful tool to correct other proxies for PCP. Continuing work will extend this study to carbonates grown on glass plates directly in the cave setting.