

Guidelines for experiments on CO₂ sequestration in peridotites based on a natural example

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Although natural examples show that olivine rich rocks react extensively with CO₂ to form ophicarbonates, it has proven to be difficult to develop practical CO₂ sequestration technologies based on olivine.

The Solund and Fensfjorden Devonian basins of SW Norway contain numerous peridotite clasts that record natural CO₂ sequestration. This situation provides a natural laboratory for studying the storage of CO₂ in basins as suggested by the IPCC [1] and determining how CO₂ reacts with peridotite in nature.

A detailed petrographic and geochemical study [2] reveals that the peridotite clasts were affected by a multi-stage alteration process. During the early stage peridotite was partly serpentinised forming the typical mesh texture with veins of serpentine surrounding compartments of relict olivine (Fo90). The olivine in the compartments is partly or completely replaced by a clay-like alteration product with a composition close to that of sepiolite (Mg₄Si₆O₁₅(OH)₂·6H₂O). Chemical analyses and mass balance calculations show that Mg was extensively removed, whereas Si remained virtually immobile. The continuous serpentine framework that persists in this alteration stage suggests that the replacement took place at constant volume.

In the most evolved stage the mineralogy is dominated by calcite and silica. Dissolution of calcite after treatment with 0.1 M HCl reveals that the silica builds up a 3-dimensional network suggesting that it was formed from the breakdown of the Mg-phases. The bulk MgO content in the overall process decreased from 40 to 2 wt%, whereas the CaO concentration increased from 1 to 35 wt%.

The findings reported here (that a clay-like mineral phase is involved in the reaction, that dominantly calcite rather than magnesite forms and that Mg is almost completely removed from the peridotite) are interpreted to be important for the storage of CO₂ in peridotites. We suggest that the most appropriate reactant for successful CO₂ sequestration is weathered peridotite (Ca-source) rather than pure olivine. Furthermore, the process must allow the removal of Mg.

[1] Metz *et al.* (2005) *Cambridge Univ. Press*, 442 pp.

[2] Beinlich *et al.* (2009) *GCA*, this volume.

Temperature-dependent formation of biogenic Cu(0) and metal sulfide colloids in flooded soil

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We recently observed that the formation of biogenic Cu(0) and metal sulfide colloids mobilized Cu, Cd and Pb in a contaminated riparian floodplain soil [1]. At the onset of soil reduction, Cu was mobilized as metallic Cu(0) crystals associated with bacteria. During sulfate reduction, two types of Cu-rich sulfide colloids formed also incorporating Cd and Pb: i) hollow particles of ~100 nm associated with bacteria likely formed by reaction of sulfide with Cu(0) and ii) dispersed nanoparticles of <50 nm likely formed by reaction of sulfide with dissolved metals.

Because temperature strongly influences the kinetics of biotic and abiotic processes, we extended our earlier microcosm experiments (done at 23°C) to 14°C and 5°C to study colloid dynamics over temperatures common for the field site (Muldenstein, Germany). Colloids isolated from the soil porewater over periods of up to 5 weeks of flooding were analyzed by transmission electron microscopy (TEM) and X-ray absorption spectroscopy (XAS).

Porewater dynamics indicated that lower temperatures increasingly delayed microbial Mn(IV), Fe(III), and especially sulfate reduction. Colloid formation was accordingly delayed, but involved the same colloid types (Cu(0), metal sulfide hollow particles and nanoparticles) as at 23°C. In contrast to 23°C, metallic Cu was not completely transformed into metal sulfide colloids. Furthermore, at 14°C and 5°C, a longer gap between the onset of Mn(IV)/Fe(III)-reduction and sulfate-reduction resulted in higher dissolved Cd levels (displaced from sorption sites by Fe²⁺ and Mn²⁺) and subsequently larger sulfide nanoparticles with higher Cd contents.

Our new results demonstrate that temperature affects metal sulfide colloid generation in a flooded wetland soil mainly via its influence on microbial reduction of terminal electron acceptors. At all studied temperatures, however, sulfate reduction led to the formation of metal sulfide colloids that have the potential to greatly enhance the release of chalcophile metal contaminants from contaminated floodplain soils into ground and surface waters.

[1] Weber *et al.* (2007) *GCA* 71, A1094.