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pH effect of Fe oxidation during abiotic and microbially mediated silicate weathering: Implications for CO_2 sequestration in subsurface storage sites

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Silicate weathering in subsurface CO_2 storage sites can significantly buffer the pH and thereby contribute to sequestration of CO_2 by shifting the carbonate equilibrium towards HCO_3^- or even by inducing carbonate precipitation. Silicate weathering occurs at low rates but might be significantly increased by microbial activity. Microbes thriving on basalt, for example, have been discovered on the ocean floor (Emerson and Moyer, 1997), where these organisms benefit from weathering basalt by using reduced iron from the rock as a source of energy. However, the partitioning of ferrous iron in different silicate mineral solutions can have an opposite effect on the pH due to oxidation of ferrous iron and precipitation of iron oxides. We address the question of how much iron oxidation coupled to basalt weathering affects the acid-base equilibrium and the ocean capability of CO_2 trapping.

For studying the mineral alteration process under abiotic conditions, we used flow-through and batch experiments at T=26° and p=1 bar. In the flow-through experiments a defined solution was pumped through columns packed with crushed minerals or rocks. Batch experiments were performed in 0.5 l closed capped polyethylene flasks and each flask contained 250 ml sterile solution and 5 grams of crushed unweathered silicate. Basalt, enstatite [$MgSiO_3$], omphacite [$(Na,Ca)(Al,Mg)Si_2O_6$], hypersthene [$(Fe,Mg)SiO_3$], quartz [SiO_2] and magnetite [$Fe_2O3*FeO$] were used as dissolving mineral phase. Artificial seawater was used as solvent with an ionic strength of about 0.7. Ca, Mg, Mn, Fe, Al, and Si were omitted from artificial seawater in order to facilitate their detection in the leachate. All systems were adjusted to pH 7.34 in case of artificial seawater (ASW) and to pH 3.55 in the case of acidified artificial seawater (aASW) by adding H_2SO_4. In blank experiments, distilled water (pH 7.29) was used. The leachate was monitored for pH; Ca, Mg and Fe content was measured using atomic absorption spectrometry and silica concentrations were measured by spectrophotometry using the molybdate blue assay. The solid phase was analysed by X-ray diffraction, scanning electron microscopy, sequential iron extraction, and petrographic thin sections. The experiments were focused on changes in the concentrations and redox state of Fe in aqueous solutions and mineral phases.

First results demonstrate that during ongoing mineral and rock dissolution, pH increases in all systems (acidified or not) to 7.0-8.2 and in the case of enstatite to 8.5-9.0. In comparison, pure quartz had no significant influence on the pH. Rates of iron leaching relative to rates of silica release were more or less congruent in both minerals with and without iron in the crystal lattice. The amount of silica leached with aASW from non-iron-containing silicates increases 3-5 times compared to ASW, whereas rates of silica releasing from iron-containing minerals are almost the same in both solutions. However, iron leaching in aASW increases about 20 times compared to normal seawater.

Results from flow-through experiments show that dissolution of various iron containing silicate minerals is more or less congruent, and that leaching and subsequent oxidation of iron imposes a significant effect on the pH. As part of this ongoing study, we will further use microbial culture experiments to compare rates and pH effects of the biotic and abiotic processes. Experimental results as well as model calculations will be used to quantitatively predict the pH effect of silicate weathering and the capacity to sequester CO_2 in subsurface storage reservoirs. These studies may also provide a basis to quantify the effect of microbially mediated silicate weathering on the global carbon cycle.

Emerson, D. and Moyer, C.L. (1997) Applied and environmental microbiology 63, 4784-92.