

Forward and reverse marine silicate weathering disentangled – an experimental study

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Marine silicate alteration (MSAlt), that includes the dissolution of silicate minerals (forward weathering, i.e. alkalinity release and CO₂ consumption) and the precipitation of authigenic clays (reverse weathering; i.e. alkalinity consumption and CO₂ release) impacts marine element cycles and global climate on geological timescales. However, due to the often simultaneously occurring mineral dissolution and precipitation reactions, quantification of element and CO₂ budgets remains challenging. Therefore, we conducted two types of batch experiments, in which marine sediments from the Sakhalin Slope (Sea of Okhotsk) were reacted with artificial seawater for 56 days. The experiments were built up to simulate two sedimentary conditions, i.e. the surface zone (without CO₂ addition) and the deeper methanogenic zone (with CO₂ addition). To identify MSAlt reactions, we used stable Si isotopes ($\delta^{30}\text{Si}$) in fluids and in situ mineral analyses using femtosecond-laser ablation MC-ICPMS measurements, in addition to major and trace element fluid and solid analyses. For both experimental runs, alkalinity, DIC, Si, B, Sr, and Li increased until a steady state was reached after 20 days. For the experiments without CO₂, $\delta^{30}\text{Si}$ increased rapidly from a starting value of $-1.0 \pm 0.2\text{‰}$ (2SD) to a steady-state value of $+2.1 \pm 0.1\text{‰}$ (2SD). For the experiments with CO₂, $\delta^{30}\text{Si}$ also increased though less rapidly from the same starting value to an average steady-state value of $+1.3 \pm 0.2\text{‰}$ (2SD). Si isotopes of bulk solids (composed of diatoms, detrital silicates and authigenic clays) in both experiments had an average value of $+0.5 \pm 0.1\text{‰}$ (2SD). We identified several successive phases of mineral reactions in dependence of the experimental time for both setups: 1) initial dissolution of diatoms and detrital smectite, 2) onset of authigenic clay precipitation, and 3) the establishment of a dynamic equilibrium between dissolution and precipitation. While this evolution of mineral reactions was seen in both experiments, the precipitation of authigenic clays was more pronounced in the experiments without CO₂. In the other experimental setup, high CO₂ concentrations caused the dominance of mineral dissolution reactions over authigenic clay formation. Quantified element budgets in these experiments will advance the interpretation of field data to disentangle forward and reverse silicate weathering in the marine environment.