

Redox and Acid Controls on Shale Chemical Weathering: Implications for Water Quality

**KATHRYN FARBER^{1,2}, LUCIEN STOLZE³, CAILEB
TRAVIER¹, BRANDY STEWART¹, ELIZABETH L
PAULUS², KRISTIN BOYE¹ AND VINCENT NOËL¹**

¹SLAC National Accelerator Laboratory

²Stanford University

³Lawrence Berkeley National Laboratory

Shale weathering is a fundamental process driving the mobilization of carbon, nutrients, and metals, with significant implications for global biogeochemical cycles. However, the impact of redox conditions and carbonic acid in regulating weathering rates remains poorly constrained, particularly in watershed systems where these parameters fluctuate due to changes in groundwater levels and microbial activity. Understanding these controls is critical for quantifying geochemical fluxes from mountainous watershed bedrocks. This study investigates how dissolved O₂ and CO₂ availability modulate shale dissolution kinetics and mineral transformations, providing insights into how redox-dynamic weathering and microbial activity pulse contributes to transport in watersheds and influences global weathering budgets.

Batch dissolution experiments were conducted under controlled dissolved O₂-CO₂ conditions to simulate natural redox and microbial activity fluctuations in mountainous watersheds. Changes in pH, major and trace element concentrations, anions, and dissolved organic carbon (DOC) were monitored over time. Kinetic results were integrated into geochemical models to parameterize weathering rates under variable redox and acid conditions. Preliminary findings indicate that concurrent O₂ and CO₂ exposure significantly enhances shale dissolution rates compared to single-gas or oxic conditions. This suggests a strong interaction between oxidative and acid-promoted weathering pathways, accelerating mineral breakdown and element mobilization beyond what may have been expected.

By quantifying shale weathering rates under variable environmental conditions, this study improves predictions of solute fluxes from terrestrial to aquatic systems, using laboratory results to inform and calibrate PHREEQC simulations. Future work will further investigate the influence of shale structure and fractures on solute transport to refine these flux estimates. Additionally, we will explore the role of organic carbon oxidation in weathering reactions and the effects of more complex, environmentally relevant porewaters, providing a quantitative understanding of mechanistic weathering dynamics. Calibrated models will eventually enhance larger-scale predictions for geochemical mass transfer and environmental responses to changing hydrological and temperature conditions.