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Controlling role of water table fluctuations and lateral flow on shale weathering in a mountainous watershed: implications for hydrogeochemical export and nitrogen cycling

LUCIEN STOLZE, DIPANKAR DWIVEDI, CARL STEEFEL, BENJAMIN GILBERT AND BHAVNA ARORA

Lawrence Berkeley National Laboratory

Presenting Author: lstolze@lbl.gov

Weathering of shale formations strongly influences carbon and nitrogen cycling and concentration of aqueous species in groundwater. Despite the significant advances in understanding the mechanisms controlling shale bedrock's alteration, the interpretation of the seasonal and long-term hydrochemical fluxes to rivers and greenhouse gases emissions to the atmosphere remains a major scientific challenge.

This study explores the coupling between the weathering of nitrogen-rich Mancos shales, the cycling of nutrients, and the groundwater flow in a pristine mountainous watershed of the East River study site, southwestern Colorado. We consider a well-instrumented and monitored hillslope-to-floodplain transect that was used for a comprehensive examination of the temporal and spatial variability of the groundwater composition and gas concentrations. At this site, alteration of shale bedrock is evidenced by the existence of a stratified weathering front characterized by the depletion of carbonate and sulfide minerals and complex trends in solid organic matter content.

We propose and develop a modeling framework to provide a quantitative understanding of the long-term weathering of shale rocks and its impact on hydrogeochemical export. In particular, we analyze the relative influence of the lateral and vertical hydrological fluxes as well as the seasonal hydrologic fluctuations on the mineral transformation and nitrogen cycling. The model simulates the exchange of gases between the atmosphere and the subsurface, the infiltration of meteoric water, a series of microbially-mediated reactions, including the cycling of nitrogen as well as the transformation of the mineral assemblage induced by dissolution/precipitation reactions.

The formulation of modeling scenarios shows that the ingress of oxygen drives the dissolution of sulfide minerals and a variety of microbially-mediated reactions. The degradation of organic matter enhances the dissolution of carbonate minerals, leading to significant emission of CO_2 into the atmosphere, and represents the primary pathway of nitrogen in groundwater. The degree of water saturation exerts a strong control on the gas fluxes and ultimately determines the extent and dynamics of weathering and the fate of nutrients. Thus, the strong water table fluctuations occurring between the dry and snow-melt periods result in rapid seasonal shifts in microbially-mediated reactions, mineral dissolution, and nutrient concentrations.