Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.20274

Biogeochemical cycling of carbon and nitrogen in the vadose zone: a multiphase and reactive transport approach

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Soil respiration and chemical weathering are recognized as major components of the global carbon (C) and nitrogen (N) cycles. Understanding the partitioning of these species at the interface between the subsurface and the atmosphere is therefore of fundamental importance for predicting feedback on Earth's climate, freshwater quality, and ecosystem functioning. Yet, accurate quantification of the C and N fluxes remains challenging due to the nonlinearity and coupled behavior associated with the physical and biogeochemical processes and the strong influence of climatic events. This study explores the cycling of carbon and nitrogen within the soil/saprolite horizon of a hillslope-to-floodplain transect underlain by C/N-rich Mancos shale and located at the East River study site, a high elevation pristine mountainous watershed of the Upper Colorado River basin. At this site, monitoring of the subsurface hydrology, aqueous and solid phase chemistry, and near-surface gas emissions reveals that snowmelt dominates the hydrologic cycle and significantly influences the mobility and fate of carbon and nitrogen. In order to quantitatively interpret the observed dynamics, we implement a process-based model that describes the partially saturated flow, the transport of reactants in multiple phases, and the key biogeochemical reactions including microbial respiration and water/rock interactions. Using the flow and reactive transport simulator CrunchFlow, we perform transient simulations to reproduce the trends of aqueous and gaseous concentrations measured along the hillslope and the flux of carbon dioxide emitted from the subsurface to the atmosphere. The simulation outcomes allow us to construct detailed biogeochemical budgets of carbon and nitrogen. We show that the soil respiration and the heterotrophic respiration of shale-associated organic matter exert key controls on the total C and N fluxes through the production of carbon dioxide and the mineralization of organic nitrogen. Furthermore, the results illuminate the importance of the coupling between microbial respiration and the dissolution of carbonate minerals as a source of inorganic carbon in groundwater. Finally, we demonstrate that water infiltration events and the associated change in moisture content modulate the seasonal and annual cycling of carbon and nitrogen by determining the spatial and temporal distribution of reactants driving the biogeochemical reactions.