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A critical look at colloid generation, stability, and transport in redox-dynamic environments: a pilot study in mountain watershed (CO)

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Mountainous watersheds are experiencing frequent hydrological disturbances with consequences for biogeochemical transformations at critical interfaces. For example, episodic wet-dry cycling at solid–water interfaces promote shifts in aqueous phase parameters (pH, oxygen, ionic strength, and ionic composition) and chemical, organic and mineral transformation of the solid phase, generating colloids (1nm–1µm), and/or influencing colloidal stability. Being typically associated with organic matter, micronutrients, and contaminants, colloids may serve as transport vectors throughout redox-affected terrestrial and aquatic systems, impacting biogeochemical reactivity downstream and the products exported to ground-/surface waters. Despite evidence that redox cycles play a significant role in generation and transport of colloids, the mechanisms, chemical composition, reactivity, and stability of generated colloids are poorly understood.

This lack of knowledge is compounded by challenges associated with the detection and characterization of colloids in natural redox environments, as well the lack of systematic experimental set-up to obtain necessary data to parameterize and validate models to account for colloids. To resolve this knowledge gap, we improved an approach combining molecular-scale characterization (*e.g.*, TEM, STXM, XAS) with field- and flow-based fractionation (FFF). FFF coupled with ICP-MS, UV-, MALS-, and zetasizer-DLS detectors separate and categorize colloid populations according to their physico-chemical compositions. Each distinct colloid populations are selected and then separately sub-sampled for deeper molecular-scale characterization.

Previously, we have examined the impact of redox changes on the generation and transport of colloids through a transect from bedrock to floodplains through a series of lab and field experiments. This work revealed that oxidative dissolution of pyrite from oxygenated water penetration through bedrock at neutral pH generates 50-100nm Fe-colloids, promoting the mobilization of nutrients and contaminants (*e.g.*, Ni and Cr). In floodplains, our results show that low sulfidation increases colloid stability of ferrihydrite, whereas higher sulfidation ($S/Fe<0.5$) generates nano-scale FeS colloids. In natural samples, however, ferrihydrite colloids are suspected to remain stable under sulfidic conditions due to the passivation by organics. Overall, we demonstrated that the colloid chemical composition drives their chemical stability through repeated redox cycling. Finally, incorporating colloidal transport highlighted in our column experiments significantly improved accuracy of model predicting floodplain biogeochemical processes.