Floodplains as biogeochemical reactors: Using multi-scale approaches to quantify hot spots and hot moments at DOE’s Rifle, Colorado field site

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The Lawrence Berkeley National Laboratory’s Sustainable Systems Scientific Focus Area (SFA) 2.0 seeks to develop a predictive understanding of how climate-induced changes in hydrology and vegetation affect watershed scale biogeochemical functioning. The Watershed” component of SFA 2.0 explores the use of multi-scale approaches to characterize terrestrial environments such as floodplain deposits at DOE’s Rifle Colorado field site across scales and compartments using geophysical geochemical mineralogical and hydrological datasets. Experiments at Rifle have long focused on stimulated biogeochemical pathways arising from organic carbon injection. Although reductive pathways have been a focus since 2002 ongoing studies are exploring oxidative pathways and their role in mediating fluxes of C N S and aqueous metals. Insights gained from such ‘stimulation’ experiments are providing insight into analogous natural biogeochemical pathways that mediate elemental cycling in the absence of exogenous carbon. Such reactions are instead mediated by endogenous pools of natural organic matter (NOM) deposited during aggradation of aquifer sediments associated with fluvial processes along the upper Colorado River corridor. Discrete lenses of fine-grained organic-rich sediments (up to 2% organic C and N) enriched in reduced species such as Fe(II) iron sulfides and U(IV) have been identified along the active margin of the floodplain through a combination of geophysical characterization approaches and drilling-recovered aquifer sediments. Referred to as “naturally reduced zones” (NRZs) these localities constitute a distinct facies type (i.e. ‘biogeofacies’) within an otherwise gravel-dominated largely NOM-deficient matrix. NRZs contain 100-fold higher U concentrations than surrounding aquifer sediments and represent 'hotspots' of seasonally intense C N S and U cycling during excursions in groundwater elevation. Along with recharge by oxic surface waters and groundwater imbibition of air bubbles within the capillary fringe during water level rise is inferred to contribute to seasonally oxic groundwater (both so-called 'hot moment' events) with the impact on redox-mediated reactions exhibiting close correspondence to those induced through intentional introduction of oxidants. Reactions induce sharp gradients in nitrate and sulfate resulting from elevated rates of nitrification and oxidation of reduced sulfur as dissolved oxygen becomes non-limiting. 7-fold increases in aqueous U are observed during this period likely contributing to U plume persistence at the site. Because NRZs contain large stores of NOM and have an outsized capacity to mediate a broad range of redox transformations such ‘hotspots’ are expected to exert influence over the redox status and biogeochemistry of floodplain deposits worldwide and are thus of broad relevance to subsurface biogeochemists.