

Title: WATERSHED CONTROLS ON URANIUM CONCENTRATIONS TIED INTO NATURAL ORGANIC MATTER AND IRON INTERACTIONS IN STREAMBEDS AND WETLANDS

Peter H. Santschi,¹ Daniel I. Kaplan², Chris M. Yeager³, Chen Xu^{1*}, Peng Lin¹, Nicole E. Patterson

¹Texas A&M University at Galveston, Galveston, TX;

²Savannah River National Laboratory, Aiken, SC

³Los Alamos National Laboratory, Los Alamos, New Mexico

Contact: (santschi@tamug.edu; xuchen66@tamu.edu)

Project Lead Principal Investigator (PI): Peter H. Santschi

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Project: Collaborative Research: Watershed Controls On Uranium Concentrations Tied To Natural Organic Matter In Streambeds And Wetlands

Project Abstract: Wetlands and hyporheic zones are critical interfacial regions with complex and seasonally-varied dynamics in hydraulic, chemical, physical and microbial properties, which result in their functioning as a sink or source of organic matter (OM) and associated contaminants, such as uranium (U). Molecular-scale understanding of the interactions between OM and U under seasonally-varied hydrological conditions (water table height, nutrients, surface water recharge and discharge inferred by ²H and ¹⁸O) in these critical zones is needed to fully resolve the regulating processes which are crucial for incorporation into watershed reactive transport models. Our **central hypothesis** is that *watershed interfacial zones, including wetlands and hyporheic zones, produce unique yet characterizable OM, with seasonal variabilities in both quantities and qualities, which are controlled by watershed hydrology/events. This seasonal, hydrological processes-driven OM dictates uranium binding and transport patterns in the long-term stewardship sites of the DOE complex.* To address this hypothesis, we are first addressing the following set of questions through field-oriented studies at the Argonne Wetland Hydrobiogeochemistry SFA field site (a U-contaminated wetland of the Savannah River Site) and accompanying laboratory experiments: 1) What is the optimal method(s) to differentially extract stable (immobile) versus mobile natural organic matter (NOM) from the same sedimentary or aquatic sample? We developed a method capable of differentially extracting immobile and mobile NOM fractions (high-molecular-weight and low-molecular-weight components for each) that are suitable for molecular-level chemical characterization via Fourier-transform ion cyclotron resonance mass spectrometry (FTICR-MS). 2) How does NOM composition impact the physical properties (e.g., hydrophobicity) underlying their aggregation/disaggregation behavior? How do diagenetic processes affect the massive occurrence of flocs in the gaining stream and the scavenging of U? We observed NOM concentrations (as calculated as the sum of carbohydrates and proteins in mg-NOM/g-particles) increasing

in the order of floc > suspended particulate matter (SPM) > bottom sediment; whereas, the protein-to-carbohydrate ratio (an index for stickiness for aggregation) was in the order of SPM > flocs > bottom sediment. Floccs contained 4 to 5-fold higher U than the stream bottom sediment. 3) What is U distribution in a dynamic watershed with relevance to groundwater-surface exchange? 4) What role does sedimentary phosphorus speciation play in U distribution in this watershed? Information from this project will identify and quantify important hydrologically driven biogeochemical processes impacting uranium at this SFA.