NOM Molecular Characterization to Better Understand Uranium Mobility in Aquifer Sediments and Groundwater

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Uranium (U) is a widespread groundwater contaminant at uranium ore milling sites as a result of natural leaching from tailings piles into the subsurface. The solubility of U is influenced by multiple factors, including redox state and the presence of other compounds within the system, such as natural organic matter (NOM). At the Department of Energy's Integrated Field Research Challenge (IFRC) site in Rifle, Colorado, subsurface sediments high in NOM are also enriched in U, suggesting that NOM has the potential to sequester U within alluvial aquifers. However, U may also be released from aquifer sediments through complexation with humic or fulvic acids. In order to address how the molecular composition of NOM influences U mobility, groundwater samples were collected from ten hydrologic locations bounding the Rifle IFRC aquifer. Dissolved organic matter (DOM) was extracted and concentrated from groundwater samples using solid phase extraction columns. Ultrahigh resolution Fourier transform ion cyclotron mass spetrometry with electrospray ionization (ESI FT-ICR-MS) was used to characterize DOM molecular form. Comparisons of DOM composition across bulk samples showed variations between the hydrologic sources that was consistent with their level of terrestrial mixing. Colorado River (CR) DOM was more enhanced in carbon heteroatoms (CHO) and depleted in sulfur heteroatoms (CHS) than was the aquifer DOM. The aquifer heteroatom compositions decreased in CHO and increased in CHS with depth and decreasing redox potential within the aquifer. DOM extracted from the Colorado River was amended to U-containing Rifle sediments to compare microbial utilization of this material to acetate-amended sediments in the context of U release/sequestration. The concentration of DOC in acetate- and CR-amended samples decreased by 70% and 56%, respectively, over a four week period while soluble U concentrations decreased 59% and 51%. This provides evidence that complex DOM can drive significant biogeochemical cycling in alluvial aquifers with implications for natural attenuation of contaminants.