Mesoscale Biotransformation Dynamics Controlling Reactive Transport of Chromium

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The interdependent influences that sediment structure and microbial communities have on transport and reduction of chromate are being investigated in various batch and microcosm systems of clay (Altamont, Calif.) and fine sand (Savannah River, S.C.) sediments. Diffusive transport of Cr(VI) has been quantified in these systems by macroscopic and synchrotron x-ray microspectroscopic methods. Indigenous microorganisms were grown in these sediments, saturated with neutral salt solutions or dilute nutrient broth (1% or 10% tryptic soy broth), prior to Cr(VI) exposure. Redox potential measurements indicated that all systems developed towards conditions favoring reduction of Cr(VI) to Cr(III) prior to Cr(VI) exposure. Cr(VI) solutions (260 to 5200 ppm) were placed in hydrostatic contact with one boundary of each sediment sample in order to simulate diffusive transport into sediment blocks from contaminant-transporting macropores. The Cr(VI) boundary reservoir was removed following 2 to 3 days of contact time to impose local conditions analogous to those expected during a short contaminant spill event. Spatially-resolved redox measurements in the sediment microcosms showed local oxidation by Cr(VI) within several mm of the exposure boundary. Spatially-resolved micro x-ray absorption near edge structure (micro-XANES) spectroscopy typically showed short Cr penetration distances, with abrupt rather than diffuse termination. Micro-XANES analysis provided direct evidence of Cr(VI) reduction to less toxic Cr(III) forms. The extent of Cr transport into sediment blocks was far less than expected by diffusion without reduction, and proportional to the boundary Cr(VI) concentration.

The microbial communities and populations in sediment microcosms were characterized with DNA fingerprints, direct counting and enrichment culturing. Intergenic Transcribed Spacer (ITS) analyses of the Altamont soil microcosm exposed to 260 ppm Cr(VI) and 1% tryptic soy broth showed that the microbial community composition in the exposure region (2 mm of the microcosm) is different from those in sediments taken from greater depth. Several populations appear only in soil that was exposed to Cr, suggesting that they are chromium resistant and that they may play an active role in Cr reduction. These results were confirmed with Denaturant Gradient Gel Electrophoresis (DGGE), where again, certain bands appeared only in fingerprints taken from soil communities that were exposed to Cr(VI). Several microbial cultures have been enriched from the sediments used in the microcosms on 10% tryptic soy broth in the presence of 100 ppm Cr(IV). These cultures will be further characterized by sequencing their 16S ribosomal genes. Direct counting of microbial populations in the sediment microcosms showed higher population densities in the outer layers for the sample exposed to 260 ppm Cr. More growth occurred in the surface layer sediment due to the availability of oxygen. Analyses of samples from other microcosms are ongoing.

These results show the important microbial and chemical heterogeneity developed from transport-limited reactions within common sediment structure. The need for measurements and models with at least mm-scale spatial resolution was demonstrated for highly nonequilibrium reactive transport in structured sediments.