BIOGEOCHEMICAL PROCESSES AND Hg CYCLING IN CONTAMINATED SEDIMENTS OF EAST FORK POPLAR CREEK, OAK RIDGE, TN (Hg SFA at ORNL)

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Site investigation and geochemical modeling provide key information on major chemical species and processes involved in mercury (Hg) biogeochemical transformations in water and sediment along a longitudinal transect of lower East Fork Poplar Creek (EFPC) in Oak Ridge, TN.

Three sites were established for regular quarterly sampling and detailed characterization of the site biogeochemistry in surface water, interstitial pore water, and creek sediments. Samples are collected from the center of the creek channel and from the creek margins. Similar measurements are being conducted on the adjacent floodplain sediments. Two of the sites (upstream – EFK22; downstream – EFK5) are on the Hg-contaminated EFPC and one is located on a nearby reference stream that receives only atmospheric Hg fallout.

Creek center channel: At both EFK22 and EFK5, 2011 water quality parameters varied little with depth and concentrations were very similar to the surface water. Porewater dissolved Hg (HgD, passes a 0.2 µm filter) showed some trends with depth yet differed between sites. At EFK22 HgD tended to decrease with depth whereas it increased with depth at the downstream location EFK5. MeHgD concentrations were lower in January and the concentration versus depth profiles were relatively uniform in the center channel at both sites in both January and August.

Creek margin: In contrast to the center channel, porewater chemistry vertical profiles exhibited distinct variations at EFK22 and EFK5, with more pronounced changes in warmer months. Changes in concentrations followed the canonical sequence of microbial terminal electron accepting processes: NO_3^- depletion, increased dissolved Mn and Fe, decreasing SO_4^{2-} concentration and increasing sulfide concentration. In intact creek sediment cores from EFK5 collected in August 2011, acid extractable Fe(II) and chrome-reducible sulfur (CRS) also increased with depth. The molar ratio of Fe(II) to sulfide in sediments averaged ~4,000. Using data from all sampling campaigns, sediment-associated MeHg is positively correlated with solid phase total Hg, Fe(II) and organic carbon content. Based on the limited results to date, there is no significant correlation between solid phase MeHg and total solid phase S(II). In general, HgD concentrations in the creek margin porewater showed little systematic variation with depth. MeHgD concentration generally exhibited a peak with depth which was shallower in August than in January. No significant correlation between MeHgD and total dissolved sulfide was observed.

Methylation ad Demethylation Potentials in Intact Cores of Creek Sediment: Coupled with the geochemical characterization of intact sediment cores, we are conducting experiments to quantify potential rates of Hg methylation and MeHg demethylation. Methylation potentials in intact sediment cores collected from EFK22 and EFK5 were significantly positively correlated with the ambient MeHg concentration but this relationship is not observed with the EFK22

floodplain samples. The EFK22 floodplain samples exhibit similar methylation potentials to the creek sediment but they have higher ambient MeHg than the creek sediments.

Data Management: Due to the volume of data generated, a data management system was adopted for archival and retrieval purposes. Standardized reporting templates, data standards, control vocabulary, and recorded metadata have been adopted. The system helps to achieve consistent reporting of field and laboratory results, including metadata, and facilitates organization and retrieval of results for analysis.