Factors influencing sediment methylmercury concentrations in a mercury contaminated creek

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East Fork Poplar Creek (EFPC), originating within the Y-12 National Security Complex (Y-12) in Oak Ridge, TN, USA, has elevated mercury concentrations as a result of historical Hg use at the facility. Stream water total mercury (HgT) concentrations decrease downstream but over this same reach methylmercury (MeHg) concentrations increase. To evaluate processes controlling the concentration of MeHg in this system, sediment cores were collected quarterly at two locations 17 km apart in EFPC between October 2010 and August 2012. MeHg sediment concentrations were most highly correlated ($p < 0.001$) with organic carbon content, Fe(II) and reduced inorganic sulfur. In sediment cores collected at the upstream site sandy, coarse grain sediment mixed with fine grain material dominated and at this site down core changes in redox sensitive parameters (Fe(II) and reduced S) were not observed. Fine grain sediments dominated in cores from the downstream site and the concentration of Fe(II) and reduced S showed increased with depth down core. At this site, mercury methylation potentials (MMP), measured using enriched stable isotopes of Hg, were higher than the upstream site. Seasonal differences in MMP, with the highest potentials measured in April, were observed in cores from the downstream site but not the upstream site. Although differences in MMP were observed between the two sites there was no significant difference in the ambient sediment MeHg concentrations. No clear differences in demethylation potential were found between sites or seasons in the creek sediment cores. Sediment cores were also collected from an ephemeral stream running through the EFPC floodplain at the upstream site and these cores were collected even when the stream was stagnant or dry. In core samples collected in October 2011 and January 2012, high concentration of MeHg were measured but methylation potentials were low suggesting MeHg production and subsequent accumulation in floodplain soils. In April 2012 ambient MeHg concentrations in the floodplain soils were lower than October and January but methylation potentials were higher suggesting either that MeHg had been transported out of the floodplain soils during the wetter winter months, conditions favoring demethylation (which was not measured in the floodplain soils) had developed, or a combination of both factors. At the downstream main channel creek site and in the floodplain ephemeral stream seasonally driven differences in temperature, which can influence microbially driven redox processes, appear to strongly influence MeHg cycling. Within the floodplain stream soil saturation status is another important factor controlling MeHg concentrations and production.