

Methylmercury and Dissolved Organic Matter Variations in a Mercury Contaminated Creek

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East Fork Poplar Creek (EFPC) in Oak Ridge, TN, USA has elevated mercury concentrations as a result of historical Hg use at the Y-12 National Security Complex (Y-12 NSC). Mercury discharge from the facility has decreased 85% since the 1980's but Y-12 NSC continues to act as a source of Hg to the system. Contaminated sediments and soils, both inside and outside Y-12 NSC, also act as a source of Hg. The objective of this work was to study the relationship between the quality and quantity of dissolved organic matter and the concentrations of inorganic and methylmercury (MeHg). Filtered (0.2 μm pore size) and unfiltered total Hg (HgT) and MeHg concentrations were measured three times in 2010 and 2011 at 7 sites along a 20-km reach of the creek. In October 2010 the concentration of filtered HgT decreased downstream from 35 to 8.4 ng/L. Unfiltered Hg varied from 125 to 300 ng/L. No decreasing trend was observed downstream likely due to variable suspended solid concentrations throughout the system. In contrast, MeHg increased from 0.10 to 0.25 ng/L and 0.15 to 0.40 ng/L in the filtered and unfiltered fraction, respectively. Filter passing HgT only constituted $7.0 \pm 4.5\%$ of the total Hg in the system while MeHg was more abundant in the filter passing fraction ($65 \pm 13\%$). The highest filtered (0.88 ng/L) and unfiltered (3.6 ng/L) MeHg concentrations were measured in an ephemeral tributary to the main creek channel draining a portion of the floodplain. Dissolved organic carbon (DOC) concentration increased downstream (1.9-3.5 mg/L) with the highest DOC concentration in the floodplain tributary (4.5 mg/L). Both total and dissolved MeHg were positively correlated with DOC concentration whereas filter passing and HgT were not correlated to DOC. Increasing specific UV absorbance (SUVA_{254}) coupled with decreasing slope ratio ($S_{275-295}/S_{350-400}$) downstream indicates an increase in molecular weight and aromaticity of the dissolved organic matter (DOM). Filter passing MeHg is positively correlated with SUVA_{254} and negatively correlated with slope ratio. These results suggest that (i) floodplain soils may serve as a source of higher molecular weight, more aromatic DOM to the creek, (ii) MeHg-particle partitioning varies with DOM quality which may have implications with respect to MeHg mobility and uptake by biota. Additional studies using ultrafiltration are being conducted to examine the distribution of mercury in different size fractions of DOM.