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Linking critical zone structure to element bioavailability across stream networks

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Trace elements are essential for aquatic life but toxic at high concentrations. Bioavailability of trace elements in streams is controlled by concentration, speciation, competing ions, and complexing ligands. Yet, how the physical structure of the critical zone regulates groundwater-surface water interactions that shape these controls remains unclear. Here, we evaluate the influence of critical zone structure on trace element bioavailability to aquatic organisms across a stream network. We use the Biotic Ligand Model to predict copper (Cu) and zinc (Zn) speciation and bioavailability in the East Fork Poplar Creek watershed in Tennessee, USA, which includes a 3rd-order stream and headwater tributaries spanning a gradient from primarily forested (low development) to urban (high development) land cover.

A cluster analysis of stream chemistry identified four stream typologies that align with flow permanence and land cover: non-perennial tributaries, low development perennial tributaries, medium-high development perennial tributaries, and the 3rd-order main stem. Flow permanence was shaped by interactions among precipitation, vegetation, urban infrastructure, and subsurface properties. Non-perennial flow ceased when vertical infiltration through permeable sediments exceeded upstream delivery, which increased with precipitation and decreased with evapotranspiration. Perennial tributaries and the main stem derived continuous flow from deeper flow paths with longer residence times. Stream chemistry reflected these contrasting flow paths along with anthropogenic inputs; that is, trace elements and solutes derived from carbonate weathering were higher in perennial streams, while solutes derived from concrete weathering and road salt increased with urban development.

Although Cu and Zn concentrations were highest in the main stem, bioavailability was limited by high concentrations of complexing ligands (CO_3^{2-}, organics) that lower free ion activities and additional ions (Na^+, Ca^{2+}) that compete for binding sites on aquatic organisms. High concentrations of dissolved organic C in less developed tributaries likely decreased metal bioavailability relative to developed tributaries. These results highlight how critical zone structure regulates trace element bioavailability across stream networks by controlling flow paths that determine concentrations of trace elements and the solutes that modify their uptake. Future work will apply this framework at the continental scale to identify patterns of potential trace element limitation or toxicity.