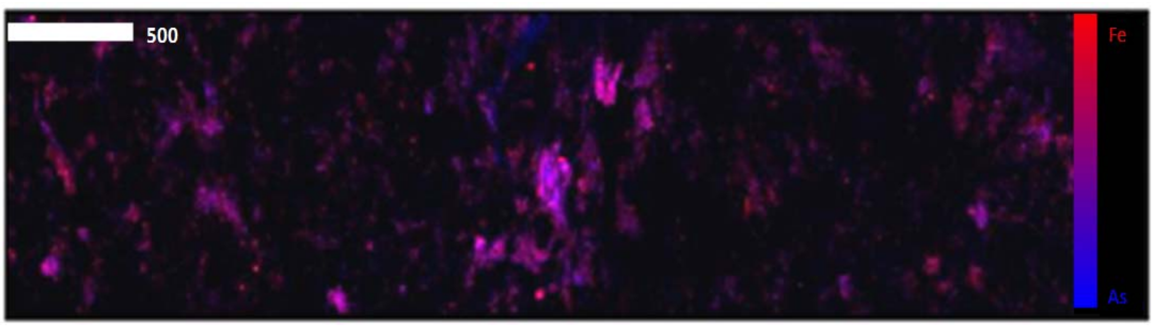
Bioaccumulation Dynamics of Arsenate at the Base of Aquatic Food Webs

Exposure to potentially toxic trace elements in freshwater systems can occur directly from water or via trophic interactions through the aquatic food web. Periphyton biofilms, which grow on rocks and sediments in freshwater streams, are an important food source at the base of aquatic food webs. These biofilms are a complex micro-ecosystem of diatoms, algae, bacteria, and fungi that can rapidly take up toxic trace elements from water and bioconcentrate these elements by hundreds to thousands of times relative to even part-per-billion concentrations in the water[1-5]. The transfer of these toxic elements to aquatic food webs and perhaps ultimately humans depends on the degree to which the periphyton concentrate the trace elements from water, and the bioavailability of the elements as they pass through periphyton-feeding higher organisms. These processes in turn depend on uptake mechanisms and chemical speciation of the trace element in the periphyton.

Recent concerns over trace elements released into rivers from coal-ash spills in Tennessee and North Carolina prompted research into trophic transfer of arsenic (As) through the aquatic food web. Arsenic is among the highest priority environmental contaminants of concern worldwide. Laboratory experiments published by Lopez *et al*. (2016) showed that periphyton rapidly takes up As from aqueous solution and bioconcentrates at 3,200 to 9,700-fold above aqueous concentrations. However, only a fraction of this accumulated As was transferred to *Neocloeon triangulifer* mayfly larvae, an aquatic invertebrate that feeds on periphyton, when it was raised for an entire lifecycle on As-enriched periphyton. Further experiments showed that assimilation efficiency of As in periphyton was modest to a variety of benthic invertebrate grazers, ranging from ~80% in the mayfly *Isonychia sp.* to ~20% in *N. triangulifer*.



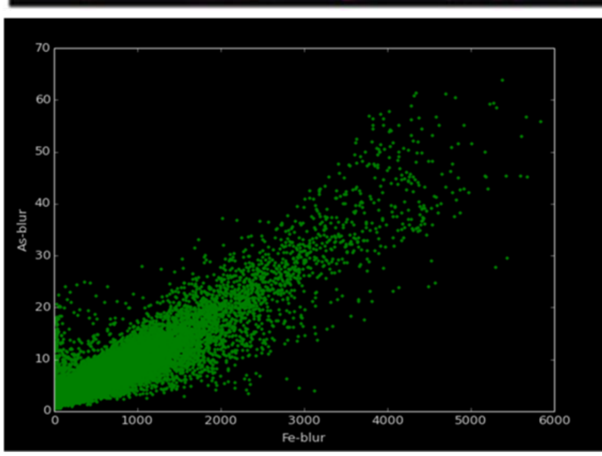


Figure. *Top*: X-ray fluorescence microprobe image showing spatial distributions of arsenic (blue) and iron (red) in a periphyton sample, with a dominance of purple to magenta areas that represent co-localization of As and Fe. The scale bar represents 500 µm. *Bottom*: Pearson correlation between As and Fe in the imaged area of periphyton (r=0.92; n=54,432).

Synchrotron x-ray fluorescence microprobe (μ-XRF) and micro-XANES imaging experiments conducted at SSRL Beam Line 2-3 showed a strong correlation of As and Fe in periphyton (see Figure). This result is consistent with strong binding and decreased bioavailability of arsenate associated with iron oxides. The research suggests that biogenic iron oxides produced by periphyton biofilms tend to scavenge arsenate from water quickly and load up the biofilms, but the bioavailability of this arsenate is also low because of its strong association with the iron oxides.

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