

Diversity of microbial arsenic transformation pathways associated with arsenic cycling in the environment

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Arsenic (As) is a naturally occurring toxic element that is widely distributed in nature. Although the concentrations of As in natural systems are generally low ($\sim 15 \mu\text{g g}^{-1}$ in soil and $\sim 10 \mu\text{g L}^{-1}$ in surface waters), the elevated levels of As have been released via natural sources (i.e. volcanic activity) and anthropogenic activities due to its increasing industrial use. As can exist in four oxidation states (-III, 0, III and V), while they are mainly found as trivalent [arsenite; As(III)] and pentavalent [arsenate; As(V)] in natural systems. Depending on its oxidation state, As exhibit different mechanisms of toxicity to microorganisms and other biota. As(III) is highly reactive with thiol containing proteins and is considered more toxic than As(V). Despite its toxicity, microorganisms have developed mechanisms to tolerate As and/or utilize the element for respiratory metabolism. Although various microorganisms have been identified to catalyze As transformation including both oxidation and reduction, we have just began to unveil the full diversity of different microbial processes associated with the redox cycling of As in the environment.

To gain insight into microbial roles in the geochemical dynamics of As, the combined geochemical, physiological and molecular biological analyses were applied to examine As-impacted environments and microcosms. Microbial populations were analyzed using 16S rDNA-based molecular approach combined with metagenomic sequencing. The presence of indigenous microbial populations capable of As transformation was examined by using both molecular approach targeting As functional genes and cultivation approach. The genes coding for arsenite oxidase (*ainA*), which catalyzes the oxidation of As(III) coupled to O₂ reduction, have been recovered from geochemically distinct geothermal habitats (pH 2.6-8) as well as the soils from mine tailing. Successful cultivation of various As(III)-oxidizing bacteria confirmed the microbial attribute in As oxidation *in situ*. In contrast, from the As impacted lake sediments and soils, diverse sequences of anaerobic arsenite oxidase (*arx*) and arsenate respiratory reductase (*arr*) genes were detected, while no *ainA* genes were recovered. The anaerobic arsenite oxidase, Arx, is known to catalyze arsenite-oxidation coupled to nitrate reduction or photosynthesis. Consistent with the molecular approach, the anaerobic arsenite-oxidizing nitrate reducer and arsenate-reducing bacteria were isolated from the lake sediments.

Our results showed that As redox metabolisms are widespread within phylogenetically and physiologically diverse bacteria, including both chemolithotrophic and organotrophic aerobes and anaerobes. This study revealed the diversity of As transformation pathways associated with geographically and geochemically distinct environments and presented the mechanisms behind microbial processes controlling the redox cycling of As.

Keywords: arsenite oxidase, arsenate reductase, microbial arsenic transformation, soil microbiology