Biosynthesis and degradation of arsenosugars

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Cyanobacteria are involved in arsenic biogeochemical cycle, and have been reported to have the ability to methylate inorganic arsenic, produce arsenosugars and arsenosugar phospholipids. In the Synechocystis sp. PCC 6803 ars operon, SSarsS is adjacent to the SSarsM gene that encodes an As(III) S-adenosylmethionine (SAM) methyltransferase. The gene product, SSArsS, contains a characteristic CX3CX2C motif which is typical for the radical SAM superfamily. The function of SSArsM and SSArsS was identified from a combination of SSarsM and/or SSarsS disruption in Synechocystis sp. PCC 6803 and heterologous expression of SSarsM and/or SSarsS in Escherichia coli. Both genes are necessary, indicating a multistep pathway of arsenosugar biosynthesis. In addition, we demonstrate that ArsS orthologs from three other freshwater cyanobacteria and one picocyanobacterium are involved in arsenosugar biosynthesis in those microbes. This study represents the identification of the first two steps in the pathway of arsenosugar biosynthesis. Arsenic accumulates in eukaryotic algae mostly in the form of arsenosugars and arsenosugar phospholipids, and those organoarsenicals are eventually released and biodegraded to maintain the dynamic balance of arsenic species in the environment. However, the specific microbiota involved in the biodegradation of arsenosugars released from the marine alga remains unknown. Our work aims to identify microbes linked to organoarsenical degradation under both anaerobic and aerobic conditions. Red alga Pyropia haitanensis was degraded under anaerobic and aerobic conditions for 28 days after being incubated in seawater with or without 1 µM arsenite under aerobic conditions for 5 days. The composition of the microbial community, total arsenic, and arsenic species were analyzed. Both total arsenic and arsenolipids were released from the algae more rapidly under anaerobic conditions than under aerobic conditions. After 28 days of aerobic incubation, the predominant arsenic species inside the algae were phosphate arsenosugars (89.1%), while dimethylarsonic acid (83.8%) was the major species under anaerobic conditions. This study offers new insights into the interaction between microbes organoarsenical degradation, contributing to our understanding of arsenic biogeochemical cycles in marine systems.

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