Supporting Information

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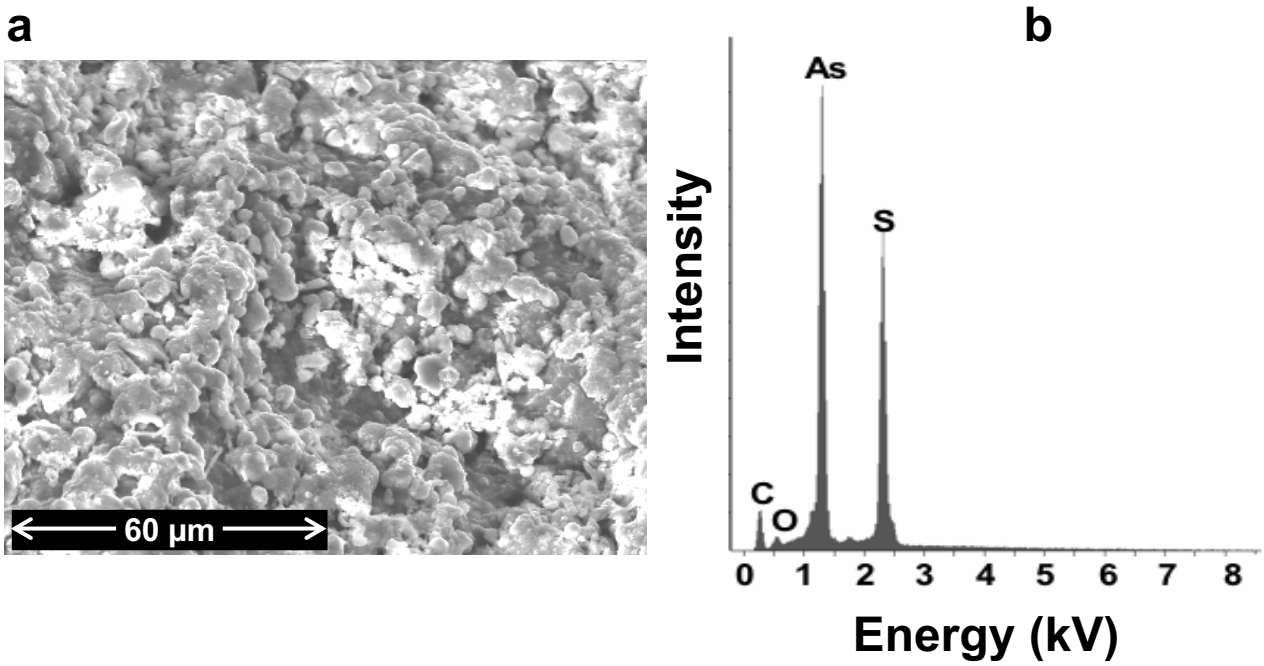

Fig. S1. Analytical examination of the orange-yellow solid phase associated with the Cyanidiales mat in Fig. 1C. (*a*) Scanning electron micrograph. (*b*) Energy-dispersive X-ray analysis. The solid-phase surface layers consist primarily of As and S at an As/S ratio of 1.1:1.3.

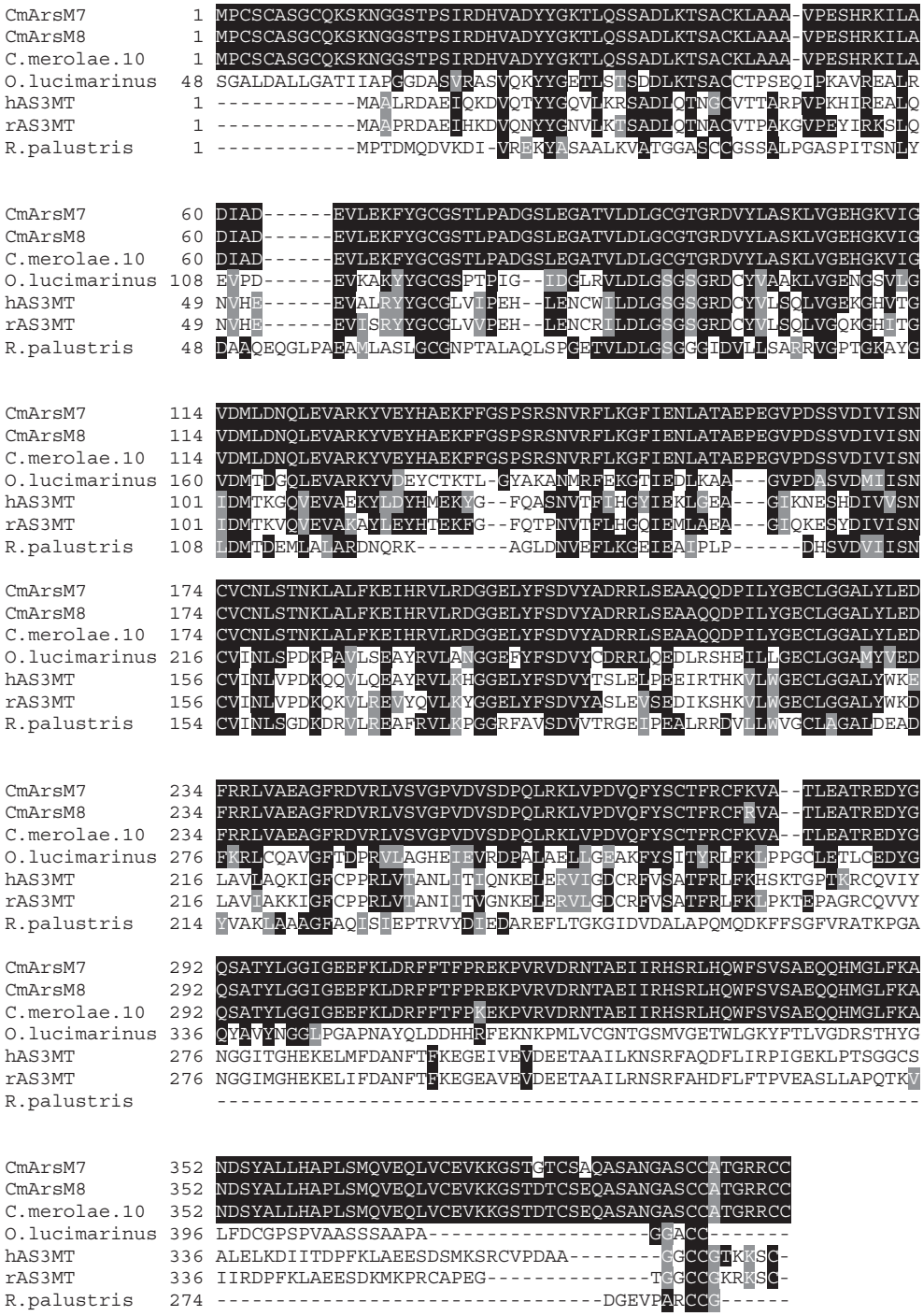

Fig. S2. Multiple alignment of ArsM homologues. *Cyanidioschyzon* strain 5508 As(III)-*S*-adenosylmethionine methyltranferases CmArsM7 (FJ476310) and CmArsM8 (FJ463403) were aligned with homologues from *C. merolae* strain 10D (AY286122), *O. lucimarinus* (ABP00263.1), human (AAI19639), rat (EDL94361) and *R. palustris* (NP_948900) (GenBank accession nos. in parentheses). The shading illustrates conservation of residues in the 3 *Cyanidioschyzon* sequences (identities in black; conserved residues in gray).

Table S1. Biomethylation of arsenite in *E. coli* expressing algal *CmarsM7*

Time, h	As(III), μM	DMAs(V), μM	TMAO, μM	TMAs(III), % total arsenic
0	15	0	0	0
3	7.9	1.1	0	0
5	1.2	7.4	0.023	5.3

The ICP-MS data from Fig. 4 were quantified by using instrumental software. The data represent the average of 2 experiments.

Table S2. Purified CmArsM7 and CmArsM8 are arsenite *S*-adenosylmethionine methyltransferases

Buffer/enzyme	Initial As(III), μM	As(III), μM	DMAs(V), μM	TMAO, μM
Control buffer	3.5	3.3	0	0
CmArsM8	3.5	0.51	4.0	0.10
CmArsM7	3.5	1.2	2.0	0.047

The ICP-MS data from Fig. 5*A* were quantified by using instrumental software. The data represent the average of 2 experiments.