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Tracing the evolution of mercury methylation reveals an early microbial "arms race"

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Phylogenetic analysis of CO dehydrogenase/acetyl co-A synthase protein family PF03599 shows the close relationship between mercury (Hg) methylating HgcA and cobalamin-dependent methyltransferases (CdhD/E) from the Wood-Ljungdahl pathway (autotrophic carbon assimilation) [1]. HgcA is less phylogenetically widespread and has a longer stem on the rooted protein tree, consistent with accelerated evolution (associated with change in function) or gene lineage extinction. The majority of experimentally-validated HgcA sequences conform to the universal tree topology, including a deepest split between archaeal and bacterial homologs. Furthermore, generally congruent topologies for HgcA/B trees support their vertical co-evolution. In combination with sister relationships of HgcA to CdhD/E, we infer the origin of Hg methylation within or before the last universal common ancestor (LUCA). Interestingly, we also found support for horizonal gene transfer (HGT) from archaea (Euryarchaeota) to bacteria (Marinimicrobia). We therefore place Hg methylation amongst Earth's earliest-evolved microbe-metal interactions, in this case resulting in synthesis of methylmercury (MeHg), a potent antimicrobial. MeHg must be exported from cells to avoid self-intoxication, possibly via formation of volatile dimethylmercury (DMHg) in the presence of aqueous or solid-phase sulfides [2, 3]. Other extracellular thiol compounds have also been shown to facilitate export and desorption of MeHg [4]. We also co-mapped the phylogenetic distribution of the *hgc* operon and *merB*, the gene encoding for alkylmercury lyase, an enzyme capable of methylmercury demethylation (i.e. detoxification). Intriguingly, with only a few exceptions, these genes were mutually exclusive across the genomes of all mapped phyla. Given their relative ages and opposing distributions, we interpret these findings as vestigial evidence for an early microbial "arms race" involving methylation/demethylation of Hg. Subsequent evolutionary radiation of *merB* may explain the observed widespread loss of *hgc* from methylating phyla under a "de-selection pressure". Other yet-to-be discovered non-*mer* detoxification mechanisms may also have facilitated *hgc* loss to explain the present day "patchy" distribution of methylators across the bacteria and archaea.

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