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Testing emergence of life hypotheses using chemical gardens simulating early Earth hydrothermal geochemistry

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Molecular hydrogen is the electron donor for the ancient exergonic reductive acetyl-coenzyme A pathway (acetyl-CoA pathway), which is used by hydrogenotrophic methanogenic archaea. How the presence of iron-sulfides influenced the acetyl-CoA pathway under primordial early Earth hydrothermal geochemistry is still poorly understood. Here, we show that the iron-sulfides mackinawite (FeS) and greigite (Fe_3S_4), which formed in hydrothermal chemical garden experiments simulating geochemical conditions of the early Archaean eon (4.0-3.6 billion years ago), produce abiotic H_2 in sufficient quantities to support hydrogenotrophic growth of the hyperthermophilic methanogen *Methanocaldococcus jannaschii.* Abiotic H_2 from iron-sulfide formation promoted CO_2-fixation and methanogenesis and induced overexpression of genes encoding the acetyl-CoA pathway. We demonstrate that H_2 from iron-sulfide precipitation under simulated early Earth (Eoarchaean) hydrothermal geochemistry fuels a H_2-dependent primordial metabolism. Additionally, we simulated the conditions of prebiotic low-temperature alkaline hydrothermal vents in co-precipitation experiments to investigate the potential of ferruginous chemical gardens to accumulate nucleic acids via sorption. The injection of an alkaline solution into an artificial ferruginous solution under anoxic conditions (O2 < 0.01% of present atmospheric levels) and at ambient temperatures, caused the precipitation of amakinite (“white rust”), which quickly converted to chloride-containing fougerite (“green rust”). RNA was only extractable from the ferruginous solution in the presence of a phosphate buffer, suggesting RNA in solution was bound to Fe2+ ions. During chimney formation, this iron-bound RNA rapidly accumulated in the white and green rust chimney structure from the surrounding ferruginous solution at the fastest rates in the initial white rust phase and correspondingly slower rates in the following green rust phase. This represents a new mechanism for nucleic acid accumulation in the ferruginous oceans of the early Earth, in addition to wet-dry cycles and may have helped to concentrate RNA in a dilute prebiotic ocean.