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Life is a verb, not a noun

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Life's emergence marks one of the last of the cascade of entropy generators that make up the evolutionary structures of the Cosmos. In far-from-equilibrium conditions, one dynamic process issues from another, and both progenitor and descendent remain coupled; e.g., convection and metabolism. Order only derives from order, not from chaos. Immaculate conception and Frankenstein-like origins of life are the stuff of myth, not science. To enable life's onset, the disequilibria and the materials capable of acting as 'free energy' converters must be locally accessible and the gradients appropriately focused. For "life is a verb, not a noun" (Gilman, 1904; Stepney, 2012). Life's job is to dissipate (the faster) whatever disequilibrium is powering it. It is, however, a construct erected on carbon scaffolds and it fundamentally has to make do with carbon from CO_2, which it must therefore hydrogenate as a foundational element of its method. And geologists since Darwin have deduced that, on rainout of the first ocean, CO_2 was available in the all-enveloping atmosphere. That is one of the initial conditions for the onset of life.

Price et al. (2017, p. 1135 in this issue of *Geology*) consider another initial condition, that "life may have emerged on early Earth in serpentinizing systems." They extend the submarine alkaline vent model for life's emergence to include basalt-hosted alkaline springs derived from meteoric waters issuing into shallow seas lapping volcanic islands. They recruit Iceland as their model, while taking into account another such site in southern New Caledonia, the Prony Bay Hydrothermal Filed (PHF). In their scenario, the entire lithospheric surface of any young, wet, rocky planet hosting a CO_2 volatisphere, whether mafic or ultramafic, provides a myriad of possible sites for life's emergence—an idea enticing to astrobiologists, and one recalling the words of Bernal: "Life, geologically speaking, consists of the interference with secondary lithosphere–atmosphere reactions so as to produce a small but ever-renewed stock of organic molecules" (Bernal 1960, p. 34).

While Price et al. acknowledge the fecundity of the submarine alkaline vent model at deep-sea ridge-flank environments such as the Lost City Hydrothermal Field (LCHF, mid-Atlantic), their islands, whether mafic or ultramafic, impose a sodium gradient to the natural proton gradient acting across precipitate membranes to aid the onset of metabolism (de Lorenzo, 2015). Price et al. model their favored environment on the Strytan Hydrothermal Field (SHF, Eyjafjord, Iceland) where maximum values of pH and temperature of the fluid feed reached 10.2 at 78 °C with a Na^+ gradient of <3:468 mM. These values are produced through interactions of meteoric waters and basalt in a ridge-flank environment precipitating saponite towers in the shallow sea occupying Eyjafjord (Table 1; Price et al., 2017). They consider that these towers make ideal "incubators" for life's birth and early evolution. The saponite is accompanied by rare calcite, and possibly stevensite and hisingerite (Table 1; Stanulla et al., 2017). In the case of the PHF, the Mg-hydroxide brucite appears to be the first precipitate, followed by a coating of Mg-carbonate, calcite, and aragonite (Table 1; Pisapia et al., 2017; cf. Okumura et al., 2016).

However, the minerals in play in the Hadean/early Archean, both in the deep sea *and* around putative basaltic islands, while also mostly hydroxides and carbonates, would have been rather different (Table 1). The main chemical sediments at that time comprised the often cherty, banded iron formations. The ferrous iron feeding these was presumably supplied from ~400 °C vents, perhaps at concentrations approaching 80 mM (Kump and Seyfried, 2005; Halevy et al., 2017). In the absence of oxygen, the iron, along with silica, is likely to have remained supersaturated until meeting with alkaline waters, of whatever derivation (Tosca et al., 2016). Indeed, that alkaline springs may have played a part in some of these developments seems inescapable. The precipitates comprised the brucite-structured mixed-valence oxyhydroxide, green rust [$\sim Fe_2(OH)_5$] in place of the true brucite [$Mg(OH)_2$] forming modern chimneys and mounds (Table 1; Arrhenius, 2003; Okumura et al., 2016; Halevy et al., 2017). And when combined with silica, greenalite would have been the Hadean/Archean phyllosilicate precipitated in place of the smectites comprising the mounds and chimneys in the Strytan field, although hisingerite appears to have been common to both the Strytan field and the banded iron formations (Table 1) (Badaut et al. 1985; Meunier et al., 2010; Tosca et al., 2016; Rasmussen et al., 2017; Stanulla et al., 2017; Price et al., 2017). Sulfides, e.g., minor mackinawite, are restricted to the Archean precipitates (Klein, 2005).

It is notable in this context that variable-valence green rust, in particular, has been viewed as critical to the emergence of life (Meunier et la 2010). Arrhenius (2003) considered the mineral as exhibiting "primitive cellular metabolic function." In this regard, it has been recently suggested that green rust and mackinawite, the objet trouvé at the ancient alkaline mounds, could have acted as the multitask protoenzymes, condensers, or nanoengines—the prebiological machinery—to enable the emergence of life (Russell and Nitschke, 2017; Muñoz-Santiburcio and Marx, 2017). Russell and Nitschke (2017) described a model whereby nitrate in the Hadean Ocean is reduced to nitric oxide, ammonia, and aminogen in the hydrated interlayers of green rust, fed with electrons from hydrothermal hydrogen. The nitric oxide contributes to the hydroxylation of methane to a methyl group, which itself reacts with formate produced in the same milieu to generate acetate,

TABLE 1. HYDROTHERMAL MINERALS PRECIPITATED IN ALKALINE CONDITIONS

MINERAL	FORMULA	SHF	PHF	LCHF	BIF
saponite	$(Ca,Na,K)_{0.2–0.3}Mg_{3.8–6.2}Si_{6.4–8.6}Al_{0.2–1.2}(OH)_4\bullet nH_2O$	✓			
stevensite	$(Ca_{0.5},Na)_{0.33}(Mg,Fe^{2+})_3Si_4O_{10}(OH)_2\bullet n(H_2O)$	✓			
hisingerite/Fe-hydroxides	$Fe^{3+}{}_2Si_2O_5(OH)_4\bullet 2H_2O$	✓			✓
brucite	$Mg(OH)_2$		✓	✓	
calcite/aragonite	$CaCO_3$	✓	✓	✓	
greenalite	$\sim(Fe^{2+},Fe^{3+})_{2–3}Si_2O_5(OH)_4,$				✓
$GR_{carbonate}$ as precursor	$\sim[Fe^{2+}{}_{6x}Fe^{3+}{}_{6(1–x)}O_{12}H_{2(7–3x)}]^{2+}[CO^{2–}{}_3\bullet 3H_2O]^{2–}$				✓
siderite	$FeCO_3$				✓
mackinawite	$FeS_{1–x}$				✓
amorphous silica/chert	SiO_2	✓			✓

Note: SHF—Strytan Hydrothermal Field; PHF—Prony Bay HF; LCHF—Lost City HF; BIF—banded iron formation.

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the target molecule of the acetyl coenzyme-A pathway. Carboxylic acids can be aminated to amino acids in the green rust nano-galleries and, thence, in theory, condensed to peptides within water lamella in nanocrysts of mackinawite (Russell and Nitschke, 2017; Muñoz-Santiburcio and Marx, 2017)—a step toward a ligand-accelerated autocatalytic metabolic pathway to a breakout denitrifying methanotrophic acetogenic metabolism (Russell and Nitschke, 2017; Muñoz-Santiburcio and Marx, 2017).

Of course, testing of ideas associating life's onset with alkaline vents of various types is a job for the laboratory. However, notwithstanding Price et al.'s (2017) caveat that the modern mounds would not survive in the geological record, a search for the vestiges of alkaline vents in banded iron formations could be rewarding. If found, they would need to be distinguished from the few known ~400 °C vents that discharged the iron into the Archean ocean. What would the precipitates be at an alkaline vent in the Hadean? To my knowledge, there is little to go on in the literature, restricted to the remark made by Goodwin that "beds of extremely carbonaceous mudrock and ironstone beds…had been disrupted by rising volatiles" in the (late Archean) Helen SVOP-IF (shallow-volcanic-platform iron formation) (Kimberley, 1989). Nevertheless, could massive developments of magnetite containing some sulfides bordered by siderite be a giveaway for such exhalative sites? Indeed, are they waiting to be identified?

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