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# Research paper Origins of building blocks of life: A review

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#### A R T I C L E I N F O

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### ABSTRACT

How and where did life on Earth originate? To date, various environments have been proposed as plausible sites for the origin of life. However, discussions have focused on a limited stage of chemical evolution, or emergence of a specific chemical function of proto-biological systems. It remains unclear what geochemical situations could drive all the stages of chemical evolution, ranging from condensation of simple inorganic compounds to the emergence of self-sustaining systems that were evolvable into modern biological ones. In this review, we summarize reported experimental and theoretical findings for prebiotic chemistry relevant to this topic, including availability of biologically essential elements (N and P) on the Hadean Earth, abiotic synthesis of life's building blocks (amino acids, peptides, ribose, nucleobases, fatty acids, nucleotides, and oligonucleotides), their polymerizations to bio-macromolecules (peptides and oligonucleotides), and emergence of biological functions of replication and compartmentalization. It is indicated from the overviews that completion of the chemical evolution requires at least eight reaction conditions of (1) reductive gas phase, (2) alkaline pH, (3) freezing temperature, (4) fresh water, (5) dry/dry-wet cycle, (6) coupling with high energy reactions, (7) heating-cooling cycle in water, and (8) extraterrestrial input of life's building blocks and reactive nutrients. The necessity of these mutually exclusive conditions clearly indicates that life's origin did not occur at a single setting; rather, it required highly diverse and dynamic environments that were connected with each other to allow intratransportation of reaction products and reactants through fluid circulation. Future experimental research that mimics the conditions of the proposed model are expected to provide further constraints on the processes and mechanisms for the origin of life.

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#### 1. Introduction

When, where, and how did life on Earth originate? These questions on the origin of life are among the biggest unsolved problems in natural science. Recent progress in geological research has provided significant constraints on the first question: time of the origin of life. The Earth was formed about 4.53 billion years (Gyr) ago through gravitational accretions of a large number of 10-km-sized objects, with the moon-forming impact being late and the most catastrophic event (Canup and Asphaug, 2001; Kleine et al., 2005; Wood et al., 2006). After the giant impact, the surface of the primitive Earth cooled rapidly from rock-melting-temperature ( $\sim$ 2000 K) to below the boiling point of liquid water

(400–500 K) on a timescale of 1–10 million years (Nisbet and Sleep, 2001: Sleep, 2010). As a consequence, a hydrosphere and continental crust were formed as early as 4.3 Gyr ago (Mojzsis et al., 2001; Wilde et al., 2001; Harrison et al., 2005). The primordial ocean was occasionally vaporized by massive meteorite impacts called the "late heavy bombardment" during the first hundreds of million years of the Earth's history (Sleep et al., 1989; Marchi et al., 2014). Liquid water is essential for life, and organic molecules are typically unstable at temperatures higher than 100 °C, therefore it is unlikely that life could have survived the bombardment period, particularly before 4.2 Gyr ago (Nisbet and Fowler, 1996; Nisbet and Sleep, 2001). Nonetheless, geochemical investigations of graphitic carbons in early Archean sedimentary rocks have indicated that life on the Earth began before 3.8 Gyr ago (Mojzsis et al., 1996; Rosing, 1999; Ohotomo et al., 2014). Geological occurrences of graphite from the Isua supracrustal belt indicate the possible existence of planktonic organisms at >3.7 Gyr ago (Rosing, 1999). If the interpretation is correct, the origin and early evolution of life would have occurred long before that. The geological evidence is

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consistent with an estimate from a phylogenetic analysis of molecular divergence among Archaea (Battistuzzi et al., 2004). A molecular clock analysis suggested that the first divergence within Archaea occurred as far back as 4.11 Gyr ago, implying even earlier dates for the last common ancestor of living organisms. Based on these multiple lines of information, the beginning of life on the Earth is inferred to be between 4.1 and 4.2 Gyr ago.

Then, where did life on Earth originate? To date, various environments have been proposed as plausible sites for life's origin, including oceans, lakes, lagoons, tidal pools, submarine hydrothermal systems, etc. But no single setting can offer enough chemical and physical diversity for life to originate. This idea was recently proposed by Stueken et al. (2013), suggesting that chemical evolution required complex interactions among diverse geochemical processes. In accord with the suggestion, Dohm and Maruyama (2014) proposed a new concept of a habitable environment. The concept, "Habitable Trinity", involves acoexisting atmosphere, water, and landmass with continuous material circulation between the three of them that is driven by the Sun. This setting is one of the minimum requirements for the emergence of life. Elements consisting of life body (C, H, O, N, and nutrients) are provided from the three components: atmosphere (C and N), water (H and O), and a landmass (nutrients). Although the presence and composition of primitive continents have been under vigorous debate (Harrison, 2009), Maruyama et al. (2013) speculated that a huge landmass had existed on the Hadean Earth (Fig. 1). It was comprised of dozens of kilometer thick anorthositic crust with local cover and dikes of KREEP (Kalium, Rare Earth Elements, and Phosphorus) basalt composition, similar to that observed on the moon. On the continental surface, serpentine-hosted hydrothermal systems distributed with localized H<sub>2</sub>-rich alkaline environments. In a pond near the geothermal field, ample and continuous supplies of nutrient elements (e.g., K and P) were provided by weathering, erosion and transportation of nutrient-enriched rocks. The shores of the pond favor dehydration reactions leading to biomolecule polymerization through wetting-drying cycles (Mulkidjanian et al., 2012a,b; Stueken et al., 2013). The geothermal fields also enabled the involvement of solar light as an energy source and diverse organic substances that were produced through atmospheric reactions or delivered by extraterrestrial objects. The proposed environment appears to include almost all geochemical situations proposed so far that favor the chemical evolution of life. Therefore, it would be worth considering whether the terrestrial environment is enough to drive all stages of chemical evolution, ranging from condensation of simple inorganic molecules (e.g., CO<sub>2</sub>) to the emergence of the earliest life. To this end, we also need to consider the last question: how did life on Earth originate?

Life is generally characterized by the following three functions: (1) compartmentalization: the ability to keep its components together and distinguish itself from the environment, (2) replication: the ability to process and transmit heritable information to progeny, and (3) metabolism: the ability to capture energy and material resources, staying away from thermodynamic equilibrium (Fig. 2; Nakashima et al., 2001; Ruiz-Mirazo et al., 2004, 2014). All these functions are operated by biopolymers such as DNA, RNA, protein, and phospholipids (Fig. 2). Phospholipids are made of two fatty acids esterified to a glycerol phosphate molecule. DNA and RNA are made of nucleosides (composed of (deoxy)ribose and nucleobases) bound by phosphodiester linkages, while proteins are made of amino acids linked together by peptide bonds (Fig. 2). It is typically assumed that these vital components were synthesized abiotically, accumulated somewhere, condensed into polymers, interacted mutually, and eventually evolved into a self-sustaining system through natural phenomena on the primitive Earth. A considerable number of laboratory simulations have been made to explore the most plausible conditions for these processes, especially for the synthesis, accumulation, and polymerization steps of organic monomers.

The aim of this review is to provide a framework for thinking about geochemical situations necessary for life to originate from

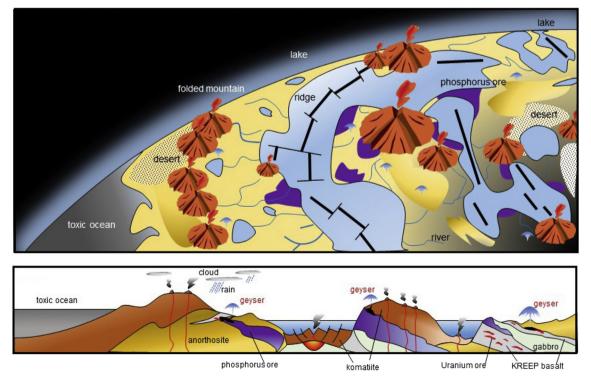


Figure 1. Schematic diagram of a Hadean surface environment proposed by Maruyama et al. (2013) and Santosh et al. (2017).

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