A global hydrothermal reactor triggered prebiotic synthesis on Earth

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Biosignatures in the rock record limit the time available for life to start on Earth to 600-800 million years[1] (4.5-3.7 Ga; Hadean-Archean). Whether the conditions for the synthesis of complex organic molecules were unique to this time or remain present today is unclear, but understanding these conditions is essential for the search of life on other planets. The outer portion of the Hadean Earth consisted of a thick mafic crust[2,3] and the upper mantle from which the crust was extracted[4]. Here we show that the recycling of the Earth's initial crust to produce the first continental crust[5–8], resulted in extreme thinning of the initial mafic crust allowing the interaction between ocean water and the upper mantle at a global scale. This global hydrothermal reactor was similar to the present-day active "Lost City Hydrothermal Field"[9], but extended on a planetary scale. The geological record indicates that the interaction between H_2O and olivine-rich rocks resulted in the production of 5-20 vol.% brucite[10–13], a key catalytic mineral for high temperature stabilisation, selection and phosphorylation of ribose[14,15]. The secular cooling of our planet[16–18], the accretion of continental crust, and deposition of sediments progressively shut down the global reactor. These processes dramatically reduced the production of brucite and the probability of synthesizing prebiotic molecules. Our results suggest that the geodynamic evolution of planets should be considered when searching for life in the wider Universe.

The period during which life originated on Earth is limited by the onset of habitability and the appearance of the first documented lifeforms[1]. In the most favourable scenario, the Earth could have been habitable as early as 4.5-4.3 Ga (Refs.[4,19]). The first lifeforms are described in the geological record at 3.7 Ga (Ref. [20]), which implies that pre-biotic molecules (PBM) must have been available and possibly abundant in this 600–800 million year time-window. Discounting an external input of life by asteroids, hydrothermal systems in an oceanic environment[9,21] or on continents, driven by magmatic activity[22] or by natural nuclear reactors[23], are potential niches for the synthesis of PBM. These systems provide the ingredients for the synthesis of the building blocks of life in the early Earth: liquid water (oceans or continental pools), a variety of gases (atmosphere and degassing) and minerals acting as catalysers[24–28].

The discovery of submarine hydrothermal vents around Galapagos[29] lead to the first hypothesis for the synthesis of PBM in high-temperature mafic-hosted hydrothermal systems[21]. This idea was later transferred to the low-temperature Lost City Hydrothermal Field (LCHF), an ultramafic-hosted alkaline hydrothermal system discovered in 2000 near the slow-spreading Mid-Atlantic Ridge[9] (Fig. 1). Low temperature (~100 ^{o}C)[30] as well as reduced and alkaline conditions[9,31] are essential for the formose reaction, and have removed some of the theoretical obstacles for a hydrothermal origin of life. Moreover, the production of a significant amount of H_2 and CH_4 and formate vital for supporting life, were considered a significant improvement with respect to the hypothesis of Ref.[21]. However, the formation of the building blocks of life is *per se* insufficient and any hypothesis for the synthesis of PBM should include selection, stabilization and phosphorylation of ribose in a natural environment[32,33].

Here, instead of starting from a defined set of chemical reactions, we take a different approach and assess which environments are potentially capable of synthesising PBM[34] in the presence of well-known catalytic minerals[26,35], were available in the early Earth. We consider that the probability of synthesizing PBM increases with the proportion of the planet in which all the essential requirements are met. Natural nuclear reactors would be punctual features as they require high-grade U ore deposits that are unlikely to be abundant in a chemically undifferentiated Hadean crust[36]. Continental hydrothermal systems are also punctual features as the supply of heat is associated with volcanic systems and distributed along belts in a discontinuous fashion. Hydrothermal systems associated with mid-ocean ridges occur along linear features. Thus, all these environments would be active on a rather limited portion of the planet. Looking back to the early Earth, after Theia's impact and the formation of the Moon at about 4.51 Ga (Ref. [37]), a magma ocean was established that cooled and degassed[38] to produce the early atmosphere in a few million years[19]. Gravitational instability of the outermost portion of the solidified magma ocean (50 vol.% olivine, 25 vol. % cpx, 20 vol.% opx, 5 vol.% plg) eventually resulted in its wholesale or incremental removal[19] accommodated by mantle ascent, its partial melting and the construction of a thick (20-40 km) crust[2,16,39]. The high degree of partial melting that formed this initial crust left a residual, olivine-rich upper mantle[2,3,16]. The geological investigation of Archean terrains suggest that the initial mafic/ultramafic crust was recycled into the mantle[2]. During recycling, the hydrated mafic/ultramafic crust would partially melt and a 10-30% (Ref.[40]) fraction of the removed material would have resurfaced as the early continental crust (Tonalite-Trondhjemite-Granodiorite; TTG)[8,41,42]. The newly formed crust was a fraction of the recycled mafic crust and the ocean floor must have been covered by much less sediments than today[43]. Thus, during this period of crustal regeneration, the potential exposure and interaction between the depleted upper mantle with water would have been more significant than at present. This, in turn, would have increased the potential for systems similar to the LCHF to develop[9,31,44–46].

At LCHF, the hydrothermal modification of the ultramafic mantle produces mainly serpentine and magnetite and, where alkaline hydrothermal fluids discharged into the ocean, additional brucite[44,45] and carbonate (Fig. 1). These hydrothermal systems (LCHF, as well as hybrid

Logatchev[47] and Rainbow[48] systems) are extremely dynamic. Multistage serpentinization generate a wide variety of niches characterised by specific pH, redox potential, temperatures, and activities of elements critical for prebiotic synthesis, distributed in space and changing with time[49–53]. Moreover, abiotic hydrocarbons and carboxylic acids have been described in hydrothermally altered mantle rocks of LCHF[54]. The following are some of the fundamental reactions occurring in hydrothermal systems hosted by mantle lithologies[55,56]. The serpentinization of olivine produces ferroan brucite, serpentine, and magnetite[12,13,44]:

$$(Mg,Fe)_2SiO_4 + H_2O = (Mg,Fe)_3Si_2O_5(OH)_4 + (Mg, Fe)(OH)_2 + Fe_3O_4 + H_2 \quad (1)$$

olivine water serpentine ferroan brucite magnetite

The formation of hydrogen is related to the amount of ferric iron in serpentine and to the moles of magnetite produced (Eq. 1)[49,55,56]. Eventual increase in temperature (e.g. magma injection) or changes in other thermodynamic variables (e.g. oxygen fugacity) destabilises ferroan brucite leading to the massive precipitation of magnetite, associated with abundant production of hydrogen:

$$(Mg, Fe)(OH)_2 + H_2O + SiO_2 = Mg(OH)_2 + (Mg,Fe)_3Si_2O_5(OH)_4 + Fe_3O_4 + H_2 + H_2O \quad (2)$$

ferroan brucite brucite serpentine magnetite

Fundamentally, the reaction between H_2 produced by hydrothermal circulation in ultramafic rocks, and CO_2 released from the mantle or magma degassing, produces CH_4 as a final product with methanediol as an intermediate reaction product (Eq. 3). The reaction of methanediol and H_2, produces formaldehyde (Eq. 4), the building block of life:

$CO_2 + 4H_2 = CH_4 + 2H_2O$ *Sebatier-type reaction* (3)

$HCOOH + H_2 \rightarrow CH_2(OH)_2 \rightarrow HCHO + H_2O$ *Formation of formaldehyde* (4)

Starting with formaldehyde and glycolaldehyde, under alkaline conditions and in the presence of cation catalysts like Mg^{2+} and Ca^{2+}, the formose reaction produces a variety of pentoses (ribose, arabinose, xylose, lyxose, ribulose, xylulose[57]). Ribose, the essential constituent of

RNA and DNA, is the least stable of the pentoses and rapidly decomposes to generate polymeric tar mixtures[14]. Selection, stabilization and accumulation of ribose, and its phosphorylation to form RNA units, are key factors to unravel prebiotic chemistry and the origin of life[33,58]. Borates and boric acid have been experimentally demonstrated to have this stabilizing effect on pentoses and to select ribose[59,60]. Moreover, phosphorus is necessary for phosphorylation, which is also assisted by borates and boric acid[61]. Because of the important role of borates, various authors have proposed different models for the accumulation of large quantities of these minerals[62], which requires differentiated and evolved continental crust and subaerial ponds undergoing desiccation. The presence of such regions with high concentrations of B and P cannot be determined with any certainty as almost all of the Hadean rock record simply does not exist. Here we propose that prebiotic chemistry did not take place in borate deposits enriched in P but was mediated by the key catalyser brucite. This mineral adsorbs large quantities of B and P (Refs.[14,63–65]), while being stable in an environment characterised by high and variable pH, a range of redox conditions, and participates in the synthesis of formaldehyde. Additionally, the reactions involving ferroan brucite and brucite modulate the release of H_2 and the availability of Mg^{2+}. These elements and conditions are all required to select ribose from the other pentoses, stabilise it to relatively high temperatures and facilitate phosphorylation which is key for the transition to self-replicating macro-molecules[14,65] (Eq. 2). While other potential catalysers could have been stabilised by the interaction between mafic crust and ocean water[66], their abundance would have been extremely limited with respect to the amount of brucite produced by interaction between water and mantle lithologies (Fig. 2). This is especially true for the olivine-rich mantle produced by high degrees of melting during the production of the early mafic crust.

We propose that during the period of crust regeneration in the Hadean-Archean, a global ultramafic reactor was active, producing copious amounts of brucite and thus PBM.

Fundamental for the plausibility of a global ultramafic reactor is the assumption that the oceanic crust was sufficiently thin to allow oceanic water to penetrate into the upper mantle and trigger the formation of brucite. To estimate the evolution of crustal thickness in time, once clement conditions were established on Earth (4.5-4.3 Ga; Refs.[19,67]), we performed mass balance calculations using a Monte Carlo approach. We rely on geological[5,68] and experimental petrological[40] constraints to define a plausible range of the parameters (Methods). The results show that due to recycling the Hadean crust progressively thins and for the largest number of simulations, reaches a minimum after around 600-700 Myrs (~3.9 Ga; Fig. 3a, b). At this time the average thickness of the oceanic crust is about 2000 m, which is significantly thinner than today's average oceanic crust (6000 m; Ref.[69]; Fig. 3c). This is in agreement with the decreasing thickness of the oceanic crust observed when large portions of the Earth crust disaggregate increasing the cooling rate of the mantle[70]. It should be noted that because we do not consider recycling of newly formed mafic and TTG-type crust, all thicknesses presented are maximum estimates. Additionally, the rate of production of new crust was likely heterogeneous, as also shown by numerical modelling[6,7]. Thus, our mass balance suggests that at around 4.2-3.9 Ga, large portions of the upper mantle were either covered by a thin oceanic crust or exposed on the ocean floor. This would have allowed the interaction between ocean water and the upper mantle[71] at a global scale, triggering the production of substantial amounts of brucite both above and below sea level (Fig. 1) and boosting the production of PBM. As these conditions were never reproduced on Earth, any process that would have isolated the mantle from the interaction with water within this optimal time window might have left the Earth a lifeless planet. At present, the upper mantle is exposed to the interaction with water only in very limited portions of our planet (0.29% of the surface; Methods). Hence, the probability of todays' Earth to synthesis PBM is vanishingly small in comparison to the Hadean-Archean. Brucite is absent on the surface of Mars, which may indicate that life as we know it on Earth may never have existed on the 'red planet'. As a result of the intimate relationship between life and the evolution of planets, the rate and sequence of geological processes should be considered when searching for life in the Universe.

Methods

We consider that the recycling of the Earth's crust once clement conditions were established (4.4-4.3 Ga; Refs.[19,67]) results in the production of TTGs[2,8,18,72]. We make no inference on the actual geodynamic process responsible for crustal recycling. We consider that the degree of partial melting of a mafic/ultramafic protolith required to produce TTGs varies between 0.1 and 0.3 (*dpm*; Ref.[40]). Estimates for the crustal thickness in the Hadean (Th_H) varies between 20 and 40 km as inferred from non-arc basalts and cratonic peridotite residues (Herzberg et al., 2010; Herzberg and Rudnick, 2012; Arndt et al., 2009). Thus, we selected this range for our calculations. On the base of these considerations, we can calculate the rate of recycling of mafic/ultramafic crust (dRR_m/dt) as a function of the rate of production of TTGs ($dTTG/dt$) and the degree of partial melting of mafic/ultramafic lithologies required to produce TTG type magmas as:

$$\frac{dRR_m}{dt} = \frac{\left[\frac{dTTG}{dt}(1-dpm)\right]}{dpm} \quad (1)$$

The fraction of volcanic rocks in Greenstone belts (V_{GB}) varies between 0.2 and 0.8, of which a fraction of 0.5 to 0.9 is represented by mafic rocks (V_m)[2]. We consider that the same proportions also apply to intrusive rocks (intrusive mafic=gabbros; I_g), but the repartition does not change the results of our calculations. On the base of these estimates, we calculate the production rate of mafic magmas produced by partial melting of the mantle (dM_m/dt) as:

$$\frac{dM_m}{dt} = \frac{dTTG}{dt} V_{GB} V_m + \frac{dTTG}{dt}(1 - V_{GB}) I_g \quad (2)$$

To compare the amount of upper mantle rocks potentially interacting with seawater in the Hadean and nowadays, we estimate the mantle presently exposed on the seafloor. Mantlederived ultramafic rocks occur along the axial valley of slow spreading ridges (spreading rates <4 cm/yr), most commonly near axial discontinuities. Ref.[73] estimated that mantle lithologies represent about 23% of the newly formed oceanic crust along slow spreading ridges. Moving off-axis (>100 km), the oceanic crust is rapidly blanketed by sediments[43] hampering the interaction between seawater and mantle rocks. Considering the total length of slow spreading ridges (31880 km), 100 km distance on each side of the ridge, and the percentage of mantle rocks exposed at the seafloor (23%), the ultramafic reactive surface represents 0.29% of the total Earth's surface.

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Figures and captions

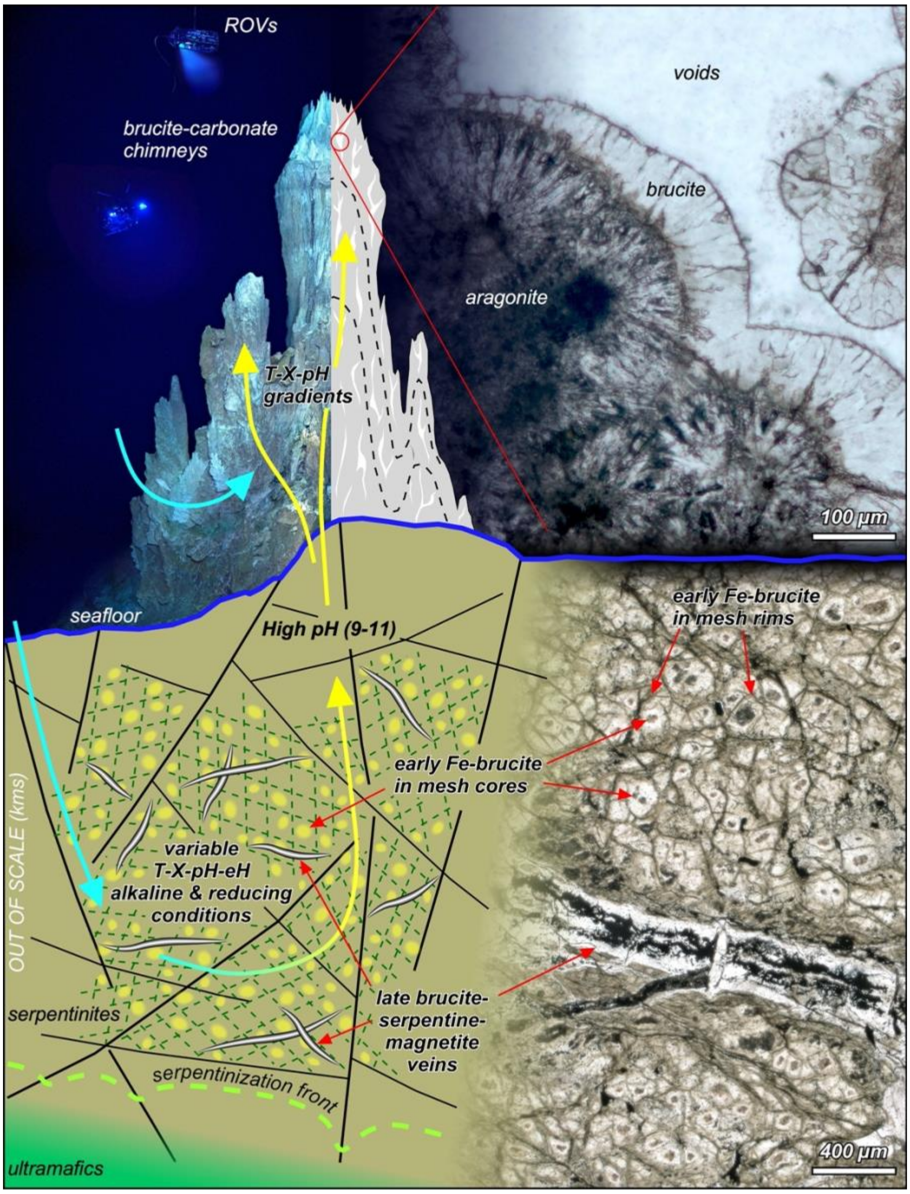

Figure 1: **An idealised sketch of one of the hydrothermal systems that constituted the global hydrothermal reactor in the Hadean-Archean.** The sketch is based on samples and geological evidence collected at the Lost City Hydrothermal Field. The widespread availability of seawater-mantle interfaces in the early Earth triggered the diffusion of brucite in a large variety of environments (subterranean, submarine and even subaerial) dominated by alkaline, reduced conditions. The residual character of Hadean mantle rocks maximised the diffusion of brucite, with its unique catalytic properties, providing an unrepeatable global scenario for prebiotic synthesis.

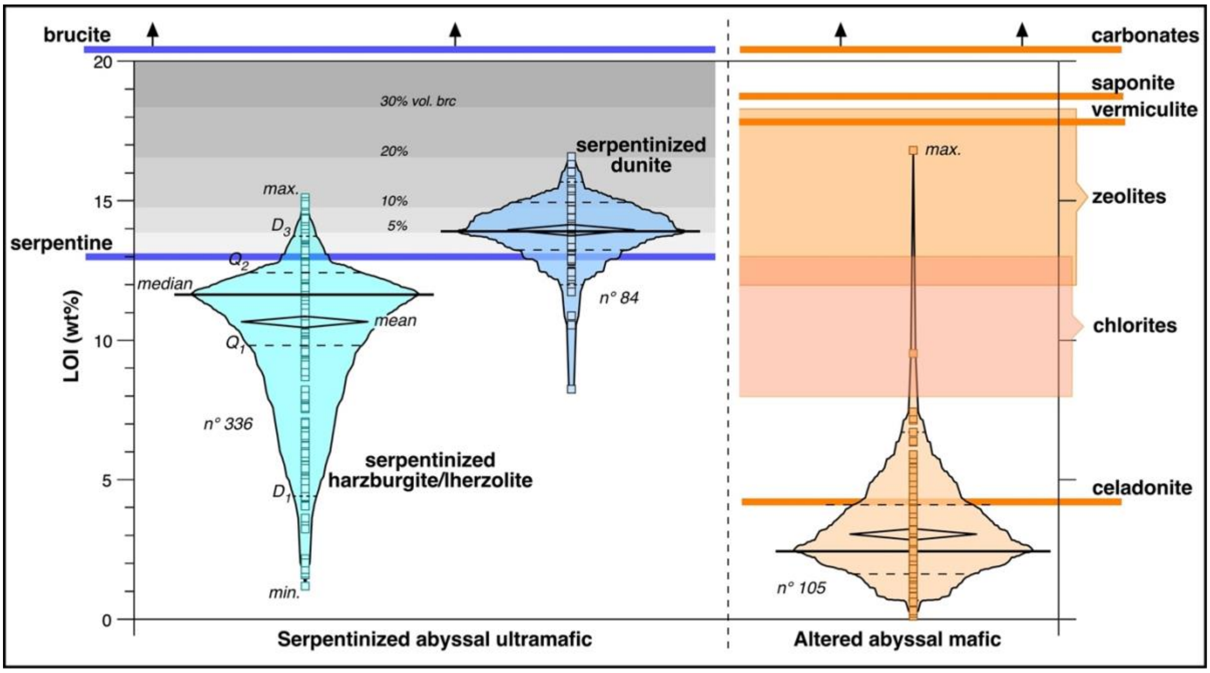

Figure 2: Box-percentile plots for abyssal serpentinized upper mantle rocks and altered basalts showing a distinct degree of alteration (visualised by loss on ignition; LOI). Mean and median of serpentinite LOI roughly overlap the LOI of serpentine minerals (with a variable brucite content) indicating the extreme efficiency of the hydration process. On the contrary, the mean and the median of altered basalt LOI are much lower than the LOI of the alteration assemblage (smectites, chlorites, zeolites and celadonite) indicating a less efficient reaction process. Furthermore, box-percentile plots for serpentinized lherzolite-harzburgite and dunites indicate that olivine-dominated systems, characteristic of the residual Hadean-Archean mantle, produce larger amounts of brucite. Data collected from Refs.[50,69,74–76].

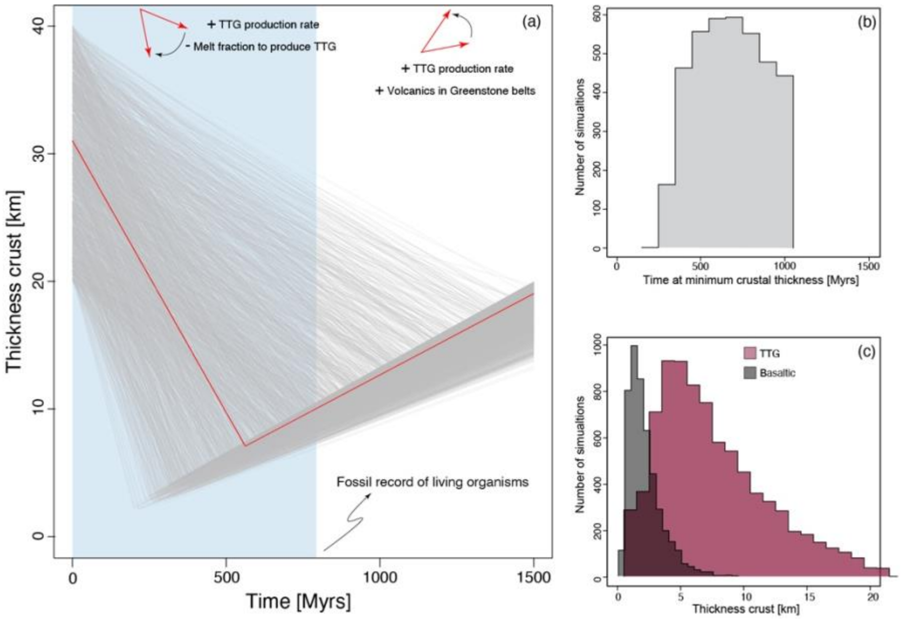

Figure 3: **Results of the mass balance calculations.** a) Grey lines show the evolution of the crust thickness in time. The red line is only to highlight one of the possible thickness-time paths. The arrows on top of the figure show the impact of the different parameters used in the Monte Carlo simulations on the rate of decrease and increase of crust thickness. b) Distribution of times at which the thickness of the crust reaches minimum values. c) distributions of the thickness of mafic and TTG crust once the total crustal thickness is at its minimum.