

Origin of the RNA world: The fate of nucleobases in warm little ponds

Ben K. D. Pearce^{a,b,1}, Ralph E. Pudritz^{a,b,c,d}, Dmitry A. Semenov^c, and Thomas K. Henning^c

^aOrigins Institute, McMaster University, Hamilton, ON L8S 4M1, Canada; ^bDepartment of Physics and Astronomy, McMaster University, Hamilton, ON L8S 4M1, Canada; ^cPlanet and Star Formation Department, Max Planck Institute for Astronomy, 69117 Heidelberg, Germany; and ^dInstitute for Theoretical Astrophysics, Center for Astronomy Heidelberg, 69120 Heidelberg, Germany

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Before the origin of simple cellular life, the building blocks of RNA (nucleotides) had to form and polymerize in favorable environments on early Earth. At this time, meteorites and interplanetary dust particles delivered organics such as nucleobases (the characteristic molecules of nucleotides) to warm little ponds whose wet-dry cycles promoted rapid polymerization. We build a comprehensive numerical model for the evolution of nucleobases in warm little ponds leading to the emergence of the first nucleotides and RNA. We couple Earth's early evolution with complex prebiotic chemistry in these environments. We find that RNA polymers must have emerged very quickly after the deposition of meteorites (less than a few years). Their constituent nucleobases were primarily meteoritic in origin and not from interplanetary dust particles. Ponds appeared as continents rose out of the early global ocean, but this increasing availability of "targets" for meteorites was offset by declining meteorite bombardment rates. Moreover, the rapid losses of nucleobases to pond seepage during wet periods, and to UV photodissociation during dry periods, mean that the synthesis of nucleotides and their polymerization into RNA occurred in just one to a few wet-dry cycles. Under these conditions, RNA polymers likely appeared before 4.17 billion years ago.

life origins | astrobiology | planetary science | meteoritics | RNA world

One of the most fundamental questions in science is how life first emerged on Earth. Given its ubiquity in living cells and its ability to both store genetic information and catalyze its own replication, RNA probably formed the basis of first life (1). RNA molecules are made up of sequences of four different nucleotides, the latter of which can be formed through reaction of a nucleobase with a ribose and a reduced phosphorous (P) source (2, 3). The evidence suggests that first life appeared earlier than 3.7 Gy ago (Ga) (4, 5), and thus the RNA world would have developed on a violent early Earth undergoing meteoritic bombardment at a rate of ~ 1 to $1,000 \times 10^{12}$ kg/y (6), which is ~ 8 to 11 orders of magnitude greater than today (7). At that time, the atmosphere was dominated by volcanic gases, and dry land was scarce as continents were rising out of the global ocean. What was the source of the building blocks of RNA? What environments enabled nucleotides to polymerize and form the first functioning RNA molecules under such conditions? Although experiments have produced simple RNA strands in highly idealized laboratory conditions (8, 9), the answers to these questions are largely unknown.

As to the sources of nucleobases, early Earth's atmosphere was likely dominated by CO₂, N₂, SO₂, and H₂O (10). In such a weakly reducing atmosphere, Miller-Urey-type reactions are not very efficient at producing organics (11). One solution is that the nucleobases were delivered by interplanetary dust particles (IDPs) and meteorites. During these early times, these bodies delivered $\sim 6 \times 10^7$ and $\sim 2 \times 10^3$ kg·y⁻¹ of intact carbon, respectively (11). Although nucleobases have not been identified in IDPs, three of the five nucleobases (uracil, cytosine,

and thymine) have been formed on the surfaces of icy IDP analogues in the laboratory through exposure to UV radiation (12). Nucleobases are found in meteorites (guanine, adenine, and uracil) with concentrations of 0.25 to 515 parts per billion (ppb) (13, 14). The ultimate source of nitrogen within them could be molecules such as ammonia and HCN that are observed in the disks of gas and dust around all young stars (15–17). A second possible source of nucleobases is synthesis in hydrothermal vents that well up from spreading cracks on young Earth's ocean floors (18). A potential problem here is the lack of concentrated and reactive nitrogen sources in these environments (18).

A central question concerning the emergence of the RNA world is how the polymerization of nucleotides occurred. Warm little ponds (WLPs) are excellent candidate environments for this process because their wet and dry cycles have been shown to promote the polymerization of nucleotides into chains possibly greater than 300 links (8). Furthermore, clay minerals in the walls and bases of WLPs promote the linking of chains up to 55 nucleotides long (19). Conversely, experiments simulating the conditions of hydrothermal vents have only succeeded in producing RNA chains a few monomers long (20). A critical problem for polymerizing long RNA chains near hydrothermal vents is the absence of wet-dry cycles.

Model: Fates of Nucleobases in Evolving WLPs

We compute a well-posed model for the evolution of WLPs, the fates of nucleobases delivered to them, and the emergence of RNA polymers under early Earth conditions. The sources of nucleobases in our model are carbonaceous meteorites and IDPs whose delivery rates are estimated using the lunar cratering record (6, 11, 21), the distribution of asteroid masses (21), and the fraction of meteorites reaching terminal velocity that are known to be nucleobase carriers (13,

Significance

There are currently two competing hypotheses for the site at which an RNA world emerged: hydrothermal vents in the deep ocean and warm little ponds. Because the former lacks wet and dry cycles, which are well known to promote polymerization (in this case, of nucleotides into RNA), we construct a comprehensive model for the origin of RNA in the latter sites. Our model advances the story and timeline of the RNA world by constraining the source of biomolecules, the environmental conditions, the timescales of reaction, and the emergence of first RNA polymers.

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¹To whom correspondence should be addressed. Email: pearce@mcmaster.ca.

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maximum bombardment model—suggests the majority of depositions occurred before 4.17 Ga. The less optimal models for WLP depositions, the LHB and minimum bombardment models, suggest the majority of depositions occurred before 3.82 Ga and 3.77 Ga, respectively.

Life Cycles of WLPs

Fig. 2 illustrates the variation of physical conditions for WLPs during the Hadean Eon, ~4.5 Ga to 3.7 Ga. Annual rainfall varies sinusoidally (24), creating seasonal wet and dry environments. The increased heat flow from greater abundances of radiogenic sources at this time (33) causes temperatures of around 50 °C to 80 °C (34). The various factors that control the water level and thus the wet–dry cycles of WLPs are precipitation, evaporation, and seepage (through pores in the ground).

In Fig. 3A, we present the results of these calculations. We select two different temperatures (65 °C and 20 °C) as analogues for hot and warm early Earths. For each of these, we examine three different environments: dry, intermediate, and wet (see Table 1 for details). The water levels in the wet environment WLPs range from ~60 to 100% full. WLPs experiencing dry states of approximately half (intermediate environment) and three quarters of a year (dry environment) only fill up to 20% and 10%, respectively. These results clearly establish the existence of seasonal wet–dry cycles.

Nucleobase Evolution in WLPs

As shown in Fig. 2, the buildup of nucleobases in WLPs is offset by losses due to hydrolysis (28), seepage (26), and dissociation by

UV radiation that was incident on early Earth in the absence of ozone (27, 36). Some protection would be afforded during WLP wet phases, as a 1-m column of pond water can absorb UV radiation up to ~95% (37). It is of particular interest that sediment, which collects at the base of WLPs, also attenuates UV radiation. Studies show it only takes a ~0.6-mm layer of basaltic sediment to attenuate UV radiation by >99.99% (38).

Nucleotide formation and stability are sensitive to temperature. Phosphorylation of nucleosides in the laboratory is slower at low temperatures, taking a few weeks at 65 °C compared with a couple of hours at 100 °C (39). The stability of nucleotides on the other hand, is favored in warm conditions over high temperatures (40). If a WLP is too hot (>80 °C), any newly formed nucleotides within it will hydrolyze in several days to a few years (40). At temperatures of 5 °C to 35 °C that either characterize more-temperate latitudes or a postsnowball Earth, nucleotides can survive for thousand-to-million-year timescales. However, at such temperatures, nucleotide formation would be very slow. Considering this temperature sensitivity, we model the evolution of nucleobases in WLPs in matching warm (5 °C to 35 °C) and hot (50 °C to 80 °C) early Earth environments. Hotter ponds evaporate more quickly; therefore, we choose rainier analogue sites on a hot early Earth to maintain identical pond environments to our warm early Earth sites.

In Fig. 3B, we focus on the adenine concentrations in WLPs from only IDP sources. The combination of spikes, flat tops, and troughs in Fig. 3B reflects the variations of adenine concentration in response to drying, balance of input and destruction rates, and precipitation during wet periods. In any environment

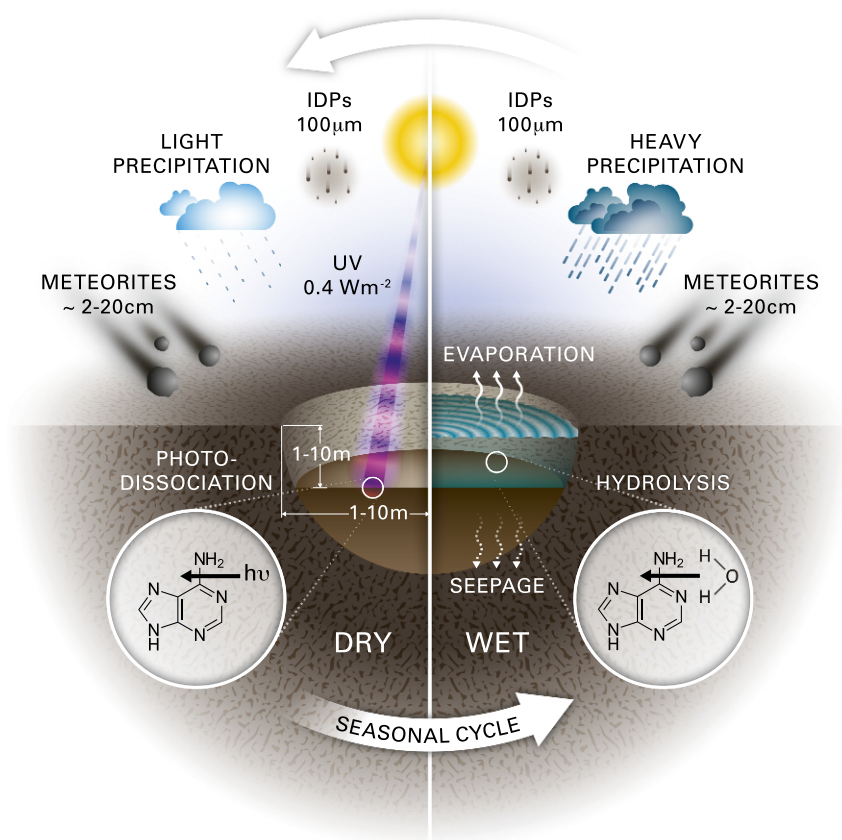


Fig. 2. An illustration of the sources and sinks of pond water and nucleobases in our model of isolated WLPs on early Earth. The only water source is precipitation. Water sinks include evaporation and seepage. Nucleobase sources include carbonaceous IDPs and meteorites, which carry up to ~1 pg and ~3 mg, respectively, of each nucleobase. Nucleobase sinks include hydrolysis, UV photodissociation, and seepage. Nucleobase hydrolysis and seepage are only activated when the pond is wet, and UV photodissociation is only activated when the pond is dry.

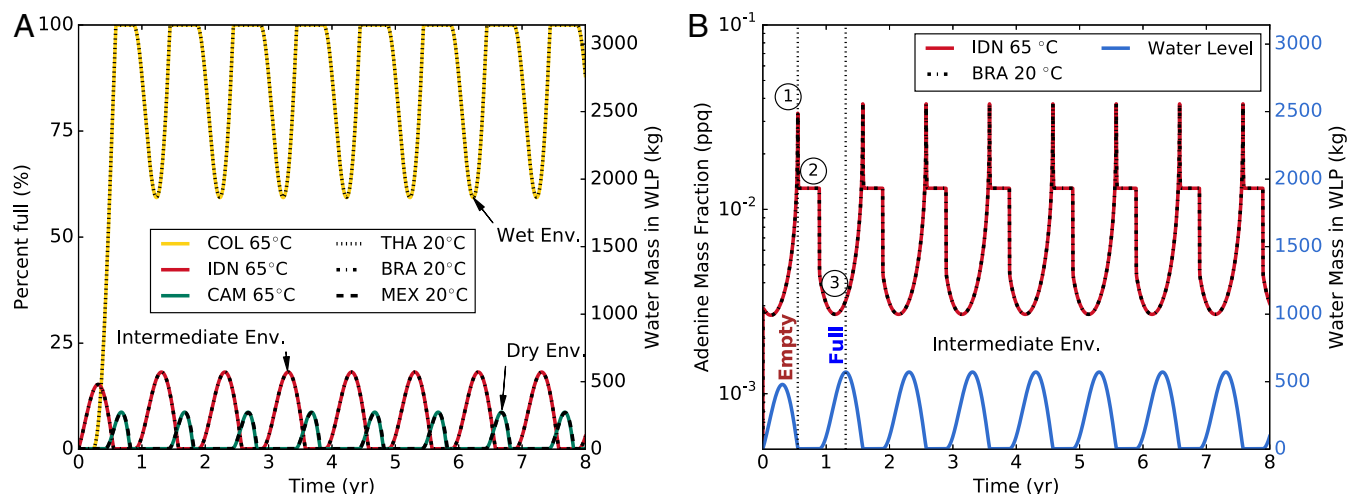


Fig. 3. Histories of pond water and adenine concentration from IDPs. (A) The change in water level over time in our fiducial dry, intermediate, and wet environment WLPs due to evaporation, seepage, and precipitation. Precipitation rates from a variety of locations on Earth today are used in the models, and represent two classes of matching early Earth analogues: hot (Columbia, Indonesia, Cameroon) and warm (Thailand, Brazil, and Mexico) (for details, see Table 1). All models begin with an empty pond, and stabilize within 2 y. (B) The red and black-dotted curves represent the adenine concentrations over time from carbonaceous IDPs in our fiducial WLPs. The degenerate intermediate WLP environment used in this calculation is for a hot early Earth at 65 °C and a warm early Earth at 20 °C. The blue curve represents the corresponding water level in the WLP, with initial empty and full states labeled vertically. Three features are present: (1) the maximum adenine concentration at the onset of the dry phase, (2) a flat-top equilibrium between incoming adenine from IDPs and adenine destruction by UV irradiation, and (3) the minimum adenine concentration just before the pond water reaches its highest level. BRA, Brazil; CAM, Cameroon; COL, Columbia; Env, environment; IDN, Indonesia; MEX, Mexico; THA, Thailand.

and at any modeled temperature, the maximum adenine concentration from only IDP sources remains below 2×10^{-7} ppb (Fig. S1). This is two orders of magnitude below current detection limits (14), making subsequent reactions negligible. The nucleobase mass fraction curves are practically independent of pond size ($1 \text{ m} < r_p \leq 10 \text{ m}$) once a stable, seasonal pattern is reached ($< 35 \text{ y}$). This is because, although larger ponds have a larger collecting area for IDPs, they have an equivalent larger area for collecting precipitation and seeping nucleobases.

Dominant Source of Surviving Nucleobases

In Fig. 4, we assemble all of these results and compare carbonaceous IDPs to meteorites as sources of adenine to our fiducial WLPs. Small meteorite fragments (1 cm in radius) are compared with IDPs in Fig. 4A. The effects of larger meteorite fragments (5 cm and 10 cm) on adenine concentration are displayed in Fig. 4B.

The maximum adenine concentration in our model WLPs from carbonaceous meteorites is 10 orders of magnitude higher than the maximum adenine concentration from carbonaceous IDPs. The reason for this huge disparity is simply that carbonaceous meteoroid fragments—each carrying up to a few milligrams of adenine—are deposited into a WLP in a single event. This allows adenine to reach parts per billion–parts per million-level concentrations before seepage and UV photodissociation efficiently remove it from the WLP in one to a few wet–dry cycles. The maximum guanine and uracil accumulated in our model WLPs from meteorites are also more than 10 orders of magnitude higher than those accumulated from IDPs (Figs. S2 and S3). A maximum adenine concentration of 2 ppm is still approximately one to two orders of magnitude lower than the initial adenine concentrations in aqueous experiments forming adenosine and AMP (2); however, these experiments only ran for an hour.

Adenine within larger fragments diffuses over several wet–dry cycles, and, during the dry phase, no outflow occurs. For fragments 1, 5, and 10 cm in radius, 99% of the adenine is released into the pond water within $\sim 10 \text{ d}$, 8 mo, and 32 mo, respectively

(Fig. S4). Only the adenine that has already flowed out of the fragments gets rapidly photodegraded; therefore, adenine within larger fragments can survive up to $\sim 7 \text{ y}$.

Thus, even though the carbon delivery rates from IDPs to early Earth vastly exceed those from meteorites, it is the meteoritic material that is the dominant source of nucleobases for RNA synthesis.

Nucleotide and RNA Synthesis

To form nucleotides in WLPs, ribose and a reduced P source must be available. Ribose may have formed and stabilized in borate-rich WLPs via the formose reaction (polymerization of H_2CO) (41). Additionally, phosphite has been detected in a pristine geothermal pool representative of early Earth, suggesting the potential availability of reduced P to WLP environments

Table 1. Precipitation models matching dry, intermediate, and wet environments on a warm (5 °C to 35 °C) and hot (50 °C to 80 °C) early Earth

Model	Environment	Analogue site	\bar{P} , $\text{m} \cdot \text{y}^{-1}$	δ_p	s_p , y
Warm early Earth (5 °C to 35 °C)	Dry	Mexico (MEX)	0.94	1.69	0.3
	Intermediate	Brazil (BRA)	1.8	0.50	0.85
	Wet	Thailand (THA)	3.32	0.91	0.3
Hot early Earth (50 to 80 °C)	Dry	Cameroon (CAM)	3.5	0.5	0.3
	Intermediate	Indonesia (IDN)	4.5	0.2	0.85
	Wet	Columbia (COL)	6.0	0.5	0.3

Precipitation data from a variety of locations on Earth today (24, 35) represent two classes of matching early Earth analogues: warm (Thailand, Brazil, and Mexico) and hot (Columbia, Indonesia, Cameroon). For example, the conditions in Mexico on a warm early Earth match the conditions in Cameroon on a hot early Earth. \bar{P} is the mean precipitation rate, δ_p is the seasonal precipitation amplitude, and s_p is the phase shift. To obtain the rate of the decrease in pond water for a given analogue site, table values are input into this equation: $dL/dt = 0.83 + 0.06T - \bar{P} [1 + \delta_p \sin(2\pi(t - s_p)/\tau_s)]$ (see SI Text).

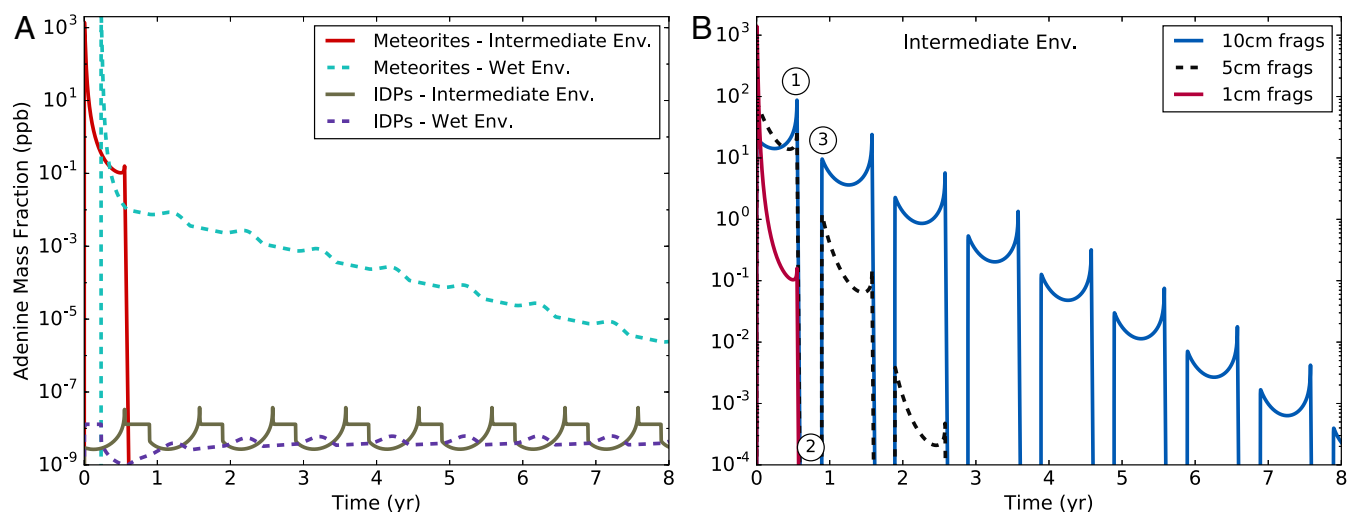


Fig. 4. Comparative histories of adenine concentrations from IDPs and meteorites. (A) A comparison of the accumulation of adenine from carbonaceous IDPs and meteorites in our fiducial WLPs. The meteorite fragments are small (1 cm), and originate from a 40-m-radius carbonaceous meteoroid. Adenine concentrations for intermediate (wet-dry cycle) and wet environments (never dry) are compared and correspond to both a hot early Earth at 65 °C and a warm early Earth at 20 °C (for details, see Table 1). (B) The effect of meteorite fragment sizes on adenine concentration. The degenerate intermediate WLP environment used in these calculations is for a hot early Earth at 65 °C and a warm early Earth at 20 °C. The fragments are either only small in size (1 cm in radius), only medium in size (5 cm in radius), or only large in size (10 cm in radius). Three features are present: (1) Adenine is at its highest concentration at the onset of the pond's dry phase. (2) Upon drying, adenine ceases to outflow from large fragments, and UV radiation rapidly destroys all previously released adenine. (3) Rewetting allows the remaining adenine within large fragment pores to continue to outflow. The U shape is due to the increase and decrease in water level. Env, environment.

(42). Only the AMP nucleotide has been experimentally synthesized in a single step involving ribose, reduced P, and UV radiation (2).

Because of the rapid rate of seepage [$\sim 1.0 \text{ mm} \cdot \text{d}^{-1}$ to $5.1 \text{ mm} \cdot \text{d}^{-1}$ (26)], nucleotide synthesis would need to be fast, occurring within a half-year to a few years after nucleobase deposition, depending on meteoroid fragment sizes. This is ample time given that laboratory experiments show that hour-to-week-long timescales are sufficient to form adenosine and AMP (2, 3).

Nucleotides, once synthesized using meteorite-delivered nucleobases, are still subject to seepage, regardless of the temperature. Therefore, nucleotide polymerization into RNA would also need to be fast, occurring within one to a few wet-dry cycles, to reduce their likelihood of seeping through the estimated 0.001- to 400- μm -sized pores at the WLP base (43). Experiments show that nucleotides can polymerize into RNA chains possibly greater than 300 monomers by subjecting them to just 1 to 16 wet-dry cycles (8).

Discussion and Conclusions

Seepage is one of the dominant nucleobase sinks in WLPs. It will be drastically reduced if nucleobases are encapsulated by vesicles (spheres of size 0.5 μm to 5 μm that form spontaneously upon hydration, whose walls consist of lipid bilayers derived from fatty acids within meteorites) (44). However, even if seepage is turned off, maximum adenine concentrations from IDPs are still negligible [$\lesssim 150 \text{ ppq}$ (parts per quadrillion)] (Fig. S5).

Also, we note that a cold early Earth [if it occurred (30)] with seasonal or impact-induced freeze-thaw cycles could also be suitable for RNA polymerization and evolution for reasons similar to

those we have analyzed for WLPs. The cyclic thawing and freezing resembles wet-dry cycles (8).

We conclude that the physical and chemical conditions of WLPs place strong constraints on the emergence of an RNA world. A hot early Earth (50 °C to 80 °C) favors rapid nucleotide synthesis in WLPs (39). Meteorite-delivered nucleobases could react with ribose and a reduced P source to quickly (less than a few years) create nucleotides for polymerization. Polymerization then occurs in one to a few wet-dry cycles to reduce the likelihood that these molecules are lost to seepage. This rapid process also reduces the likelihood of setbacks for the emergence of the RNA world due to frequent large impacts, also known as impact frustration (45). Sedimentation would be of critical importance as UV protection for nucleobases, nucleotides, and RNA (27, 46, 47).

The mass delivery model providing the most WLP depositions indicates that the majority of meteorite depositions occurred before 4.17 Ga. The first RNA polymers would have formed in WLPs around that time, prefiguring the emergence of the RNA world. This implies that the RNA world could have appeared within ~ 200 to 300 million years after Earth became habitable.

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