

1 **The co-evolution of life and organics on Earth: expansions of energy**
2 **harnessing**

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23 Abstract: The organic matter was absent prior to planetesimal formation (4.6 Gyr) but
24 at present abundant in planetary environments. The aim of this study was to combine
25 information about the organic inventory of the Earth, which is accompanied by the
26 evolution of life. A variety of available free energy sources, including geochemical
27 energy, sunlight, oxygen and fire have supported life evolution. In the meantime these
28 energy sources have mediated the diversity and complexity of living organisms and
29 resulted in a concomitant increase in the diversity and complexity of organic matter,
30 including microbial-, plant-, fire-, and human derived organics. The change of the
31 diversity and complexity of organic matter (microbial-, plant-, fire- and
32 human-derived organics) have in-return significantly influenced Earth's carbon
33 cycle, planetary climate and ecosystems. Overall, energy harnessing and
34 conservation of life entwined and expanded the evolutionary histories of life and organic
35 molecules on the planet. Considering the key role of organics on the stability of the
36 oxygen level of the atmosphere, temperature, the tectonic rise of continents, and global
37 habitability, the changing characters of organics over geologic time had an important
38 shaping influence on Earth's geochemical cycles.

39 Keywords: organic matter; life; energy harnessing

40 **1. Introduction**

41 Life is processes of generating reduced organic compounds from carbon dioxide as well
42 as the harnessing of environmental energy. Over the course of Earth history, the harnessing of
43 free energy by organisms has had a dramatic impact on the geosphere, including minerals and

44 organics (Dietrich et al., 2006; Grosch & Hazen, 2015; Judson, 2017), shaped the whole
45 trajectory of life evolution. As a direct consequence of a coevolving geosphere and biosphere,
46 the Earth's crust has changed greatly over billions of years. The origin and evolution of
47 organic compounds on the planetary environment are compelling because of their potential
48 role in the origin of life and sustaining microbial communities (Lazcano & Miller, 1996;
49 McDermott et al., 2015; Schönheit et al., 2016). Carbon lies at the heart of carbon-based life
50 forms and provides unparalleled potential for earth evolution. The origin of life is
51 inextricably linked to the behavior of carbon (Hazen, 2019). The evolution of organics is
52 coupled with the evolution of life, which is expanded with a variety of available free energy
53 sources (Judson, 2017). Collectively, these linkages have mediated the generation and
54 transformation of soils and sediments. Here we review the origin and evolution of organics
55 on Earth, and their relationship with diversification and expansion of energy utilization and
56 with biological and geological development.

57 **2. The prebiotic organics**

58 Earth accreted 4.56 billion years (Gyr) ago from largely homogeneous material (Judson,
59 2017; Hazen et al., 2013). With the dissipation of thermal energy produced by compaction,
60 radiation, and impacting meteorites, the Earth cooled. About 4.4 Gyr ago patches of a rocky
61 scum had solidified and eventually separated into core, mantle, and crust (Mojzsis, 2010).
62 Water vapor condensed as rain and formed early oceans and seas (Wilde et al., 2001; Rosing
63 et al., 2006; Hazen et al., 2013). The early ocean was a reservoir of inorganic elements, and
64 also a reservoir of potential free energy in the form of protons. Before the emergence of life

65 on early Earth at ~3.8 Gyr (Dodd et al., 2017), these prebiotic organics were either
66 synthesized abiotically on the Earth itself or synthesized extraterrestrially and then delivered
67 to the Earth (Hayes et al., 1967; Dalai et al., 2016). A wide range of organic compounds
68 including amino acids, monocarboxylic acids, sugars, nucleobases, and membrane-forming
69 lipids have been synthesized in prebiotic conditions simulation experiments (McCollom,
70 2013). Questions remain, however, concerning whether the conditions that allow synthesis of
71 these compounds in the laboratory accurately simulate those that might have been present on
72 the early Earth (McCollom, 2013; Dalai et al., 2016). High concentrations of the reactants,
73 water pH and ambient temperature are of central importance in experimental abiotic synthesis
74 of organics. The extreme environments (highly acidic condition) of early Earth presented
75 severe limitations with respect to their potential for prebiotic chemistry because of stability
76 and synthetic pathway issues associated with temperatures and pH (Bada, 2013). It has been
77 claimed that autocatalytic metabolic-like reactions can overcome these limitations (Huber &
78 Wächtershäuser, 1998). The micro-conditions in the hydrothermal systems supposedly could
79 drive abiotic syntheses of organics (McCollom & Seewald, 2007; McDermott et al., 2015).
80 Prebiotic syntheses could have taken place in a variety of geochemical environments that
81 may have existed on the primitive Earth, although this has never been demonstrated using
82 plausible geochemical conditions (Box 1). Highly reducing fluids such as deep-sea
83 hydrothermal fluids have the potential for abiotic reduction of dissolved inorganic carbon to
84 produce organic compounds (Shock, 1990; Shock & Schulte, 1998; Seewald et al., 2006;
85 McDermott et al., 2015). There is also increasing evidence that supports an abiotic origin for
86 CH₄ and other low-molecular weight reduced organic compounds in ultramafic-hosted

87 hydrothermal systems (Charlou et al., 2002; McCollom & Seewald, 2007; Proskurowski et al.,
88 2008). Given the scarcity of suitable abiotic regime the yield prebiotic organics on the early
89 Earth would have been very small (Lollar et al., 2002).

90 Besides the abiotic synthesis of organic molecules on the young Earth driven by various
91 energy sources such as UV radiation in sunlight, cosmic rays, X-rays, hypervelocity impacts,
92 volcanic eruptions with lightning, geothermal heat, and redox gradients, the total inventory of
93 organics would have included exogenous sources (the interstellar medium, interplanetary dust,
94 asteroids, comets, meteorites) (Dalai et al., 2016; Kwok, 2016; Sahai et al., 2016; Sandford et
95 al., 2016). It has long been speculated that Earth accreted prebiotic organic molecules from
96 impacts of carbonaceous asteroids and comets during the period of 4.5 Gyr to 3.8 Gyr ago
97 (Chyba et al., 1990; Chyba & Sagan, 1992; Botta & Bada, 2002) because the exogenous
98 delivery has showered the Earth (Pizzarello & Weber, 2004). Polyhydroxylated compounds
99 (such as sugars, sugar alcohols and sugar acids) are formed under interstellar conditions via
100 photolysis of small molecules (e.g. CO, NH₃ and H₂O) and are therefore present in meteorites
101 (Agarwal et al., 1986; McDonald et al., 1996; Cooper et al., 2001). The carbonaceous
102 component of interplanetary dust could be up to 50 wt% (Ehrenfreund & Charnley, 2000;
103 Dalai et al., 2016). This dust material has been reported to contain simple aliphatic, aromatic
104 compounds, macromolecular polyaromatic hydrocarbons (Ehrenfreund & Charnley, 2000;
105 Dalai et al., 2016), amino acids and other organic compounds (Cooper et al., 2001) that are
106 vital to the origin of life. Tens of thousands of tons of interplanetary dust particles enter the
107 Earth's atmosphere annually, and the rate may have been much greater on early Earth (Kwok,
108 2016). It was estimated that Earth was also accreting intact cometary organics at a rate of at

109 least 10^9 to 10^{10} g per year at 4.5 Gyr (Chyba et al., 1990). Organics delivered from space
110 comprising as much as perhaps 10% of the Earth's modern biomass by weight (Sephton, 2002;
111 Schönheit et al., 2016) estimated that about 1.0×10^{21} mol of reduced carbon were probably
112 delivered to the surface of Earth by asteroids (4.4 - 3.8 Gyr) (Catling et al., 2001; Hayes &
113 Waldbauer, 2006).

114 **3. Geochemical energy**

115 Organics underpin the co-evolution of Earth's geosphere and biosphere. Organics likely
116 played critical roles in the origin of life, and, in return, life has played a symbiotic role in the
117 production and cycling of organics. The emergence of life on Earth gave rise to a source of
118 organics in both abundance and diversity. Life began very early, before 3.8 Gyr (Des Marais,
119 2000; Nisbet & Sleep, 2001). Two main theories, based on heterotrophic versus
120 chemoautotrophic metabolisms, have emerged to account the origin and early evolution of
121 life (Ferry & House, 2006; Herd et al., 2011; Schönheit et al., 2016). Theories for autotrophic
122 origins posit that the first cells satisfied their carbon needs from CO (Say & Fuchs, 2010;
123 Fuchs, 2011; McDermott et al., 2015). While the heterotrophic theory proposes that life arose
124 from an "organic soup" of diverse preexisting molecules which were delivered from space or
125 abiotically formed (Lazcano & Miller, 1999; Bada & Lazcano, 2002). Regardless of the
126 chemoautotrophic or heterotrophic origins, organisms had evolved to take advantage of the
127 available energy to fuel their proliferation and to produce new organic matter.

128 At this time in Earth history, oxygen was at trace levels (Canfield et al., 2006), so the
129 first ecosystems must have existed in an anoxic world and their activities were driven by

130 anaerobic metabolisms (Canfield et al., 2006; Judson, 2017). The proposed emergency of life
131 under anoxic geothermal environments implies that life started not as a planetary but as a
132 local phenomenon. It was reported that metabolisms of early anaerobic ecosystems were
133 probably 2–3 orders of magnitude less active than the present biosphere (Des Marais, 2000).
134 Given these factors and the probable limits on accessing the most limiting chemical
135 compounds, various ecosystems most likely existed in relative isolation (Canfield et al.,
136 2006).

137 Noting that the energy budget of Earth places strict constraints on fluxes of basic
138 components required for chemoautotrophic life, life was unable to influence the Earth's
139 carbon cycle in any significant way in the absence of photosynthesis (Rosing et al, 2006).
140 Geochemical models (Bergman et al., 2004; Berner, 2009) suggest that the productivity of the
141 biosphere before it was powered by sunlight harvested through photosynthesis, would have
142 been at least a thousand times less than it is today (and maybe one million times less).
143 Combined continental reservoirs of organic carbon probably grew very slowly through the
144 Earth history and were still negligible before 3.5 Gyr ago (Godderis & Veizer, 2000; Canfield
145 et al., 2006). Owing to the low productivity of the non-photosynthetic early biosphere, its
146 initial influence upon the life-energy-organic dynamic would have been small (Canfield et al.,
147 2006; Sleep & Bird, 2007; Judson, 2017).

148 **4. Sunlight**

149 The greatest energy source in the surface environment of the Earth is sunlight. Today the
150 average solar energy flux to Earth surface is 340 W/m^2 (Rosing et al., 2006). The early Sun

151 was fainter and solar luminosity was probably a quarter to a third less than the present day (ca
152 250 W/m^2 at 4.0 Gyr; Sagan & Chyba, 1997; Nisbet & Sleep, 2001).

153 It is reported that by ca. 3.7 Gyr (Fig. 1) (Rosing, 1999; Pecoits et al., 2015; Nutman et
154 al., 2016), photosynthetic organisms emerged to harness the energy in sunlight to drive
155 chemical reactions. When the biosphere developed photosynthesis, living organisms acquired
156 the ability to absorb solar energy and convert a fraction of it into chemical free energy
157 (Rosing et al., 2006). Photoautotrophs acquired the ability to build up gradients in chemical
158 potential, rather than just exploiting existing gradients, as was the fate of their
159 chemoautotrophic predecessors. With evolution of photosynthesis, energy resources available
160 for lives became several orders of magnitude larger than that available from
161 oxidation-reduction reactions (chemoautotrophic primary production) associated with
162 hydrothermal activities (Des Marais, 2000; Rosing, 2005; Rosing et al., 2006). The
163 development of photosynthesis allowed life to escape the hydrothermal setting (Nisbet &
164 Sleep, 2001). Energy harvested from sunlight, therefore, enhanced the rates of autotrophic
165 carbon fixation, and carbon burial in anoxic environments. The primary productivity of the
166 photosynthetic world was estimated to be 10,000 times higher than those of
167 non-photosynthetic ecosystems (Sleep & Bird, 2008; Summons & Hallman, 2014), although
168 the rates would have been significantly lower than the present (Summons & Hallman, 2014).
169 The earliest photosynthetic organisms performed anoxygenic photosynthesis, and were
170 dependent on mineral sources as electron donors, but relieved the energy constraints to
171 perform reduction of organic compounds (Olson & Blankenship, 2004; Rosing et al., 2006).
172 The genesis of photosynthesis had irreversible consequences for Earth surface environments

173 whether it was oxygenic or anoxygenic.

174 Estimates suggest that anoxygenic phototrophs increased the flux of carbon through the
175 biosphere and the most active ecosystems were probably driven by the cycling of Fe^{2+} , with
176 the oxidation of Fe^{2+} yielding potentially the highest rates of primary production (Canfield et
177 al., 2006). The importance of hydrogen as an early fuel for anoxygenic photosynthesis has
178 also been emphasized by Olson, (2006), and may have been sufficiently abundant in the early
179 Earth to drive CO_2 reduction. Other dynamic ecosystems would have also been driven by the
180 microbial cycling of sulfur and nitrogen species, but these would have been considerably less
181 active in comparison with those based on iron and hydrogen as electron donors to reduce CO_2
182 (Canfield et al., 2006). For all the ecosystems mentioned above, the production rates of
183 organics were considerably less than those of today. The primary production rates of total
184 carbon at 3.8 Gyr ago were estimated as 2.8×10^{14} mol yr^{-1} . Organic carbon accounted for
185 14% of the total carbon. This primary production estimate is 14 times lower than present rates
186 (4.0×10^{15} mol yr^{-1}) (Canfield et al., 2006). Considering that prokaryotic life was flourishing
187 and presumably widespread in the biosphere, organics must have been completely
188 microbially derived, which produces more labile organic matter with high H/C (the degree of
189 aliphatic character) and low O/C or (O + N)/C (the degree of polar character) ratios (Qiu et al.,
190 2014) such as lipid-, protein-, and amion sugar-like products (Fig. 2) (Brocks et al., 1999;
191 Grannas et al., 2006; D'Andrilli et al., 2015).

192 **5. Oxygen**

193 As the third most energetic oxidant, oxygen reduction provides the largest potential

194 source of energy per electron transfer, except for the reduction of chlorine and fluorine
195 (Catling & Claire, 2005). Given the much less abundance of both chlorine and fluorine
196 (several orders of magnitude less than oxygen) and their high reactivity with organics, their
197 energy cannot be harnessed for life. On account of chemical sinks (such as reduced
198 geothermal outflows and rock weathering) greatly exceeded the abiotic sources of oxygen
199 (UV photolysis of water) ambient oxygen levels were insignificantly low (approximately
200 10^{-14} of present atmospheric O_2 levels) before oxygenic photosynthesis arose (Rosing et al.,
201 2006; Buick, 2008). With the evolution of more advanced oxygen producing photosynthetic
202 pathways, life became independent of both energy and reducing power derived from mineral
203 substrates (Rosing et al., 2006). Geologic evidence suggests that oxygenic photosynthesis
204 originated before 2.8 Gyr (Fig. 1) (Des Marais, 2000). At present cyanobacteria are the most
205 numerous ($\sim 10^{27}$) among all organisms performing oxygenic photosynthesis (among green
206 plants, phytoplankton and cyanobacteria) (Catling & Claire, 2005). Cyanobacteria raised
207 oxygen levels in the atmosphere ($> 10^{-3}$ present atmospheric O_2 level) by around 2.32 Gyr.
208 This planetary change to atmospheric O_2 levels is referred to as the Great Oxidation Event
209 (GOE) (Kopp et al., 2005; Buick, 2008). Oxygenic photosynthesis was clearly well
210 established by this time (Schirmer et al., 2013).

211 The stratospheric ozone layer seems to have been created at 2.3 Gyr (Goldblatt et al.,
212 2006). The formation of the ozone layer was facilitated by O_2 levels rising to 1-3% of present
213 levels; at these levels, photolysis of oxygen (yielding reactive oxygen radicals) was
214 sufficiently frequent for an ozone layer to be produced, which shielding life from short-wave
215 UV-radiation (200-300 nm) and was suitable for life to expand on the continents (Goldblatt et

216 al., 2006). Beyond the energetic limitations, defined by the availability of oxygen, the only
217 limiting factors for life from the environment became the availability of bio-essential
218 elements (Rosing et al., 2006). The increase in the oxygen content of the atmosphere and
219 ocean driven by photosynthesis increased chemical weathering rates (Shields, 2007), which
220 in turn increased nutrient (e.g. phosphorus, partly nitrogen, and iron) availability (Shields,
221 2007; Kump, 2010). The far-reaching impact of the GOE cannot be emphasized enough: it
222 changed Earth's history by enabling the evolution of aerobic life, an explosion in the
223 biosynthesis of organics and this underpinned the opportunity for organics to be generated
224 and to proliferate in large quantities on a planetary scale.

225 Oxygenic photosynthesis is by far the most efficient mechanism for harvesting solar
226 energy (Rosing et al., 2006). The GOE changed Earth's history by enabling the evolution of
227 aerobic life (eukaryotes) and the emergence of the lineage that would eventually produce land
228 plants. On Earth, aerobic metabolism provides about an order of magnitude more energy for a
229 given intake of food than anaerobic metabolism (Judson, 2017). As a consequence of
230 energetic limitations, life without O₂ as a strong electron acceptor, well mixed in the
231 atmosphere and the surface ocean, could not grow large and complex (Lane & Martin, 2010).
232 On account of prohibitively low growth efficiencies and energetic limitations anaerobes do
233 not grow beyond the complexity of uniseriate filaments of cells (Schulz & Jørgensen, 2001;
234 Catling & Claire, 2005). The oxygenated atmosphere and ocean enabled the evolution of
235 more complex life (Payne et al., 2009; Dahl et al., 2010; Kump, 2010). The maximum body
236 size of organism has increased by 16 orders of magnitude since emergence of life; this
237 transition occurred via two discrete steps (Payne et al., 2009). The first was the emergence of

238 eukaryotic cell (~1.9 Gyr) and the second was eukaryotic multi-cellularity (0.6-0.45 Gyr).
239 These two steps coincide or slightly postdate with increases in atmospheric oxygen levels
240 (Payne et al., 2009). The evolution of Earth's biota is intimately linked to the oxygenation of
241 the atmosphere and the oceans (Dahl et al., 2010). This atmospheric oxygenation correlates
242 with the diversification and radiation of vascular plants on the continents (Gensel, 2008) and
243 the oxygenation of the oceans correlates with the expansion of large predatory fish (Bambach,
244 2002). This evolution significantly enhanced the burial of reduced carbon and was accompanied
245 by the accumulation of organic matter (Fig. 1).

246 There exists a striking temporal overlap between the atmospheric oxidation and the rise
247 of the continents (Dietrich et al., 2006; Rosing et al., 2006). Continent shaping was probably
248 associated with burial of organic matter fixed by oxygenic photosynthetic organisms under
249 sediment eroded from the new blocks of crust (Des Marais et al., 1992; Dietrich et al., 2006).
250 The rifting of large continental plates on the global scale probably promoted the development
251 of extensive anoxic basins favorable for organic preservation (Des Marais et al., 1992),
252 promoted the burial of refractory plant material (e.g., lignin, cellulose, and of other refractory
253 organic compounds) (Berner, 2009).

254 The emergence of larger, and less easily degradable organic molecules related to
255 eukaryotic diversification thus enhanced the burial of organic matter and its diversity. One of
256 the most unique and pervasive biological characteristics of organic matter in terrestrial
257 environments is the predominance of sources from vascular plants (Oades, 1993). For
258 example, during the Carboniferous period (360-300 Ma), oxygen in atmosphere rose to

259 between 30 and 35% (Berner et al., 2003; Hsia et al., 2013), coinciding with the appearance
260 giant vascular plants, fern-dominated forests (Shear, 1991). Fern are lignin rich plants that
261 contain > 40% of lignin than modern plants (~20%) (Robinson, 1990), and this lignin was
262 difficult to decompose until organisms like fungi evolved and effective degradation occurred
263 until 200 million years after fern plant emergence (Robinson, 1990). The rise of ligniferous
264 plants and low lignin breakdown (due to the rare or absence of lignolytic organisms)
265 contributed to increased terrestrially derived organic matter burial through inhibited
266 decomposition (Robinson, 1990; Berner et al., 2003). The spread of vascular plants in the
267 terrestrial environment increased the diversity of organic matter, including plant-derived
268 polysaccharides such as cellulose and phenolic compounds such as lignin (Benner et al., 1984;
269 McLatchey & Reddy, 1998). These organic compounds are characterized by less labile, more
270 recalcitrant chemical nature with H/C <1.5 (Fig. 2) (D'Andrilli et al., 2015).

271 **6. Fire**

272 To trigger wildfire, all of three conditions must be met (Scott & Glasspool, 2006). Firstly,
273 a source of ignition—such as lightning strikes, meteor strikes and volcanic activity.
274 Throughout the Earth history, these have been abundant. Lightning is the pre-eminent source
275 of heat for the ignition of fossil wildfires. Lightning strikes occurred more than 1.4 billion
276 lightning strikes per year owing to its global frequency (44 ± 5 strikes/s) (Christian et al.,
277 2003), of which an appreciable number ignite wildfires. Secondly, sufficient amount of
278 oxygen became present in the atmosphere. Assuming current atmospheric pressure, at least
279 16% oxygen is the minimum concentration in order for plants to ignite and for fire to be

280 self-sustaining (Belcher & McElwain, 2008; Belcher et al., 2010). For most of Earth's history,
281 oxygen levels have been lower than this threshold until 0.35 Gyr (Scott, 2000; Scott et al.,
282 2013). Thirdly, fire requires fuel. The earliest land plants (embryophytes) evolved from
283 charophycean green algal ancestors (Steemans et al., 2009) at approximately 0.47 Gyr
284 (Berner, 2009). The appearance of vascular plants on land occurred around 0.42 Gyr ago,
285 although they were tiny and leafless (Lenton, 2001; Banks et al., 2011). All three conditions
286 were met and fire activity has begun to influence the Earth system and the cycling of organic
287 matter.

288 The evolution of plants increased the atmospheric oxygen concentration, contributing to
289 increase the amount of oxygen for fire formation. In the meantime, plants provide the fuel for
290 fire. Fire activity would be globally distributed, even in wetter climatic areas as when oxygen
291 reaches levels >30%, fire can be sustained (Scott & Glasspool, 2006). The Carboniferous
292 period was characterized as a 'high-fire' world due to elevated levels (35%) of oxygen
293 (Berner, 2006; Glasspool & Scott, 2010). A diverse vegetation provided a major and
294 extensive fuel resource although vast swamps were present on the continents (Berner, 1999).
295 Significantly enhanced fire activity continued during the Cretaceous (145–65 Ma) (Belcher et
296 al., 2010), and is hypothesized to be associated with the rise of angiosperms during this
297 period (Bond & Scott, 2010; Bond & Midgley, 2012).

298 Fire has had both geological and biological impacts on ecosystems. Fire regimes drive
299 the evolution of plant traits, such as thick bark (Bond et al., 2005; Pausas & Keeley, 2009;
300 Keeley et al., 2011); the initial spread of flowering plants (Bond & Scott, 2010); faunal

301 abundance and diversity such as ants (Moreau et al., 2006); shape biomes (Crisp et al., 2010,
302 2011; Scheiter et al., 2012); affect soils quality and nutrient cycling such as the carbon,
303 oxygen and phosphorous cycles; promote biodiversity (Bond & Keeley, 2005). Due to the fire
304 integration to ecosystem function and maintenance (Keeley & Rundel, 2005; Edwards et al.,
305 2010), pyrophilic grasslands and savannas such as C₄ grasslands expanded and replaced
306 woodlands (Keeley & Rundel, 2005; Hoetzel et al., 2013).

307 The emergence of fire in terrestrial environment likely had a profound effect upon the
308 compositions and dynamics of organic carbon. Furthermore, fire contributes new material to
309 the Earth—pyrogenic carbon or fire-derived organic matter (partly charred organic matter
310 including black carbon, charcoal and soot) (Lenton, 2013; Judson, 2017). Glinka, (1914)
311 described that “there was almost no soil profile in which charcoal particles did not occur in
312 the upper horizon” (Bird et al., 2015). In modern peats, charcoal may constitute 4% of the
313 total volume. In the Carboniferous, charcoal represented more than 20% of dead organic
314 matter (Scott et al., 2013). It was recently estimated that 3-5 million km² of the Earth surface
315 are burned by wildfires annually (Jones et al., 2019) and approximately 116-385 Tg/yr of
316 pyrogenic carbon are now produced globally by fires (Santin et al., 2016). Pyrogenic carbon
317 can represent a significant proportion of total organic carbon in the environment: ranging
318 from 2% to 60% of the total soil organic carbon in terrestrial systems (Singh et al., 2012;
319 Reisser et al., 2016). Santin et al. (2016) provided a global assessment of pyrogenic carbon
320 fluxes. Accounted for 8 to 27% of the annual production of pyrogenic carbon were inputted to
321 oceans from rivers and most of pyrogenic carbon was deposited on the continental shelf
322 (Santin et al., 2016). These reports indicate that pyrogenic carbon is a significant component

323 in both terrestrial and oceanic carbon storage (Preston & Schmidt, 2006). Moreover,
324 pyrogenic carbon has a condensed aromatic structure with low H/C and O/C ratios; it is
325 therefore recalcitrant (D'Andrilli et al., 2015). It has been established that pyrogenic carbon
326 can persist in soils and sediments for millions of years and thus it plays an important role in
327 the global carbon inventory of the Earth and fluxes between reservoirs over long time scales
328 (Schmidt & Noack, 2000; Forbes et al., 2006; Scott, 2010). Human activities that suppress
329 the production of pyrogenic carbon have significantly disturbed the pyrogenic carbon cycle
330 (Bowman et al., 2011; Andela et al., 2017).

331 The advent of anthropogenic fire was a revolutionary event in Earth history because fire
332 technology has significantly influenced the biosphere over the last 10,000 years (Box 2)
333 (Raupach & Canadell, 2010). Fire gave protection, extended the range of food, and expanded
334 adaptation to different environments on Earth (Froestad & Shearing, 2017). Fire plays a
335 pivotal role in the clearing of forests to create permanent fields with the development of
336 sedentary agriculture-based societies during the Holocene (Bowman et al., 2013). During the
337 late Quaternary humans have dramatically altered fire regimes around the globe, which is
338 largely dependent on fossil fuels, both directly and indirectly. The production and existence
339 of pyrogenic carbon underpin the significant perturbations of the carbon cycles both, on long
340 (million year) (Berner, 1999; 2003) and on short (thousand year) timescales. The application
341 of fire by humans, especially the fossil fuel burning, has accelerated both long- and
342 short-term carbon cycles through anthropogenical alternation of carbon fluxes, the increase of
343 CO₂ in the atmosphere, and global warming (Berner, 1999). Organic aerosols such as soot
344 from fire smoke in Earth's atmosphere is an important contributor to global climate change

345 by absorbing heat and warming the air (Bond et al., 2013; Berner, 2003; Johansson et al.,
346 2018).

347 Life is a process of harnessing energy to maintain states far from thermodynamic
348 equilibrium, leading to an energy flow through the biosphere (Raupach & Canadell, 2010).
349 Any reduction of an energy source could cause a corresponding contraction in the biosphere
350 and drop in the rate of global organic matter burial. In the traditional “big five” mass
351 extinctions oceanic anoxic events (due to the worldwide reduction of oxygen) have coincided
352 with four of these mass extinctions, especially, the end-Ordovician (Zhang et al., 2009), the
353 Late Devonian (Goddéris & Joachimski, 2004), the end-Permian (Wignall & Twitchett, 1996;
354 Grice et al., 2005), and the end-Triassic (Isozaki, 1997). Thus, rapid declines in atmospheric
355 O₂ have been proposed to have a major influence upon mass extinction events. For example,
356 in the most severe extinction (loss of as much as 95% of all species on Earth) that occurred in
357 the Late Permian (~251 Mya) (Benton & Twitchett, 2003; Grice et al., 2005; Chen & Benton,
358 2012), the oxygen level (at 30% or more in the Carboniferous period) fell dramatically to
359 13% in the late Permian and the early part of the subsequent Triassic (Lane, 2007). One
360 factor in the last mass extinction (end-Cretaceous) may be the prevention of sunlight from
361 reaching the surface of the Earth due to the dust, soot or aerosols in the atmosphere ejected by
362 the Chicxulub asteroid impact (Kring, 2007).

363 The sudden mass mortality of the terrestrial and marine organisms irreversibly
364 reorganized the global carbon cycle (Caplan & Bustin, 1999; Berner, 2002). A drop in burial
365 fluxes for global organic carbon is coincident with the abrupt biota change at the time of late

366 Permian (Berner, 2005) and the end-Cretaceous mass extinctions (D'hondt et al., 1998). Such
367 a reduction in the organic flux could have been a natural consequence of the ecosystem
368 reorganization that resulted from the mass extinctions.

369 **7. Conclusions**

370 The amount and proportion of pyrogenic carbon in total organic carbon on Earth are
371 expected to increase, which changed the diversity and complexity of organic matter on Earth.
372 The change of the diversity and complexity of organic matter (microbial-, plant-, fire- and
373 human-derived organics) have in-return significantly influenced Earth's carbon cycle,
374 planetary climate and ecosystems. The long-term organic carbon cycle has dominant
375 influenced the levels of atmospheric oxygen and carbon dioxide over a multimillion-year
376 time scale. The levels of atmospheric carbon dioxide and oxygen were mainly mediated via
377 weathering of organic matter on the continents, the burial of organic matter in sediments, and
378 the thermal breakdown of organic matter at depth. The increased oxidation of organic carbon
379 to carbon dioxide by weathering and thermal breakdown results in the O₂ consumption and
380 CO₂ production. The burial of organic matter in sediments leads to an increase in atmospheric
381 O₂ and a decrease of atmospheric CO₂ due to a net excess of photosynthesis over respiration
382 (Berner & Caldeira, 1997; Berner, 1999). Current burning of fossil carbon results in a
383 decrease of atmospheric O₂ by about 2 ppm per year (Keeling and Manning, 2014). The
384 fluctuation of atmospheric levels of O₂ and CO₂ would change the planetary climate,
385 temperature, precipitation (enhanced atmospheric CO₂ leads to a warmer and wetter climate
386 via the greenhouse effect), resulting in the evolution of biology. An integrated view of

387 assessing the role of the change of diversity and complexity of organic matter is required. A
388 full understanding of the role of pyrogenic carbon on planetary climate and ecosystems can
389 provide us with new opportunities for mitigating climate change.

390 If the development of life-energy-organic dynamics on other life-planet systems have
391 paralleled those on Earth (i.e. microbial-, plant-, fire-derived and anthropogenic organics),
392 then it follows that by analyzing the type of organics in their soils or rocks the evolution of
393 life-energy-organic histories/dynamics can be speculated upon (Hazen, 2019). For example, if
394 the soils or meteorites from another planet have only simple organic molecules it might be
395 inferred that the planet is in a prebiotic (or early life) stage. In contrast, where the soils or
396 meteorites from a planet have plant-derived or pyrogenic carbon, like Mars (Lin et al., 2014),
397 there exist implications for a possibly complex biosphere being present. Given, the entwined
398 evolution of life, energy utilization and organics there exist the possibility to evidence the
399 development of life elsewhere in the universe though assessment of organic matter profile
400 and the fingerprint they provide of biotic diversification.

401

402 **Acknowledgements**

403 This project was financially supported by the Natural Science Foundation of China (No.
404 41571130062), National Key Research and Development Program of China
405 (2017YFD0800303).

406

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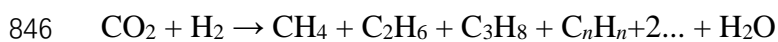
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833 **Box 1 Abiotic Synthesis of Organic Compounds in Hydrothermal**

834 **Environments**

835 Since the discovery of deep-sea hydrothermal systems in the late 1970s **Error! Reference**
836 **source not found.** (Corliss et al., 1979), there has been keen interest in the origin of life in
837 these environments. The concept that deep-sea hydrothermal systems as sites of abiotic
838 organic synthesis is based largely on their strongly reducing chemical environments. Based
839 on geological observations as well as theoretical and experimental constraints, the theory
840 received support as having the potential for abiotic synthesis of organic compounds within
841 hydrothermal environments (Charlou et al., 2002; Martin et al., 2008; McDermott et al.,
842 2017). The abiotic formation of organic compounds in geologic systems involves the abiotic
843 reduction of dissolved inorganic carbon ($\Sigma\text{CO}_2 = \text{CO}_2 + \text{HCO}_3^- + \text{CO}_3^{2-}$) to organic
844 compounds by dissolved H_2 produced by serpentinization, which can be expressed by the
845 following reaction (McCollom & Seewald, 2007):



847 The synthesis of CH_4 and organics from H_2 and CO_2 releases energy. Geothermal energy is
848 transferred into chemical energy in the form of organic compounds. These reactions take
849 place readily on the Earth. Geochemistry thus offered fresh chemical, energetic, and
850 thermodynamic perspectives on biochemical origins (Martin, 2012).

851

852 **Box 2 Fire and Hominins**

853 The use of fire is a defining feature of humans with reliable records of fire use by
854 hominins dated at 1 million years (Myr) ago (Berna et al., 2012; Bowman et al., 2013). The
855 habitual use of fire for preparing food about 0.400 Myr (Roebroeks & Villa, 2011; Sandgathe
856 et al., 2011) supported the larger human brains (Carmody & Wrangham, 2009) and relatively
857 small gut given body size (Milton, 1999). Fire was a central evolutionary force and cooked
858 diets tend to provide more energy for growing energy-expensive brains (Roebroeks & Villa,
859 2011).

860 With the harnessing of fire and the technological explosion, fire was replaced by the
861 internal combustion engine. Considering that most of the energy used by human beings
862 comes from the combustion of fossilized organic matter it might be asserted that humans
863 have become the most important evolutionary force on the planet (Palumbi, 2001)
864 considering most of the energy used by human beings comes from the combustion of
865 fossilised organic matter. Industrial-scale use of energy flows from fossil carbon have
866 significant effects on the climate, atmosphere, hydrosphere, and on global biogeochemistry
867 (Gillings et al., 2015). These changes have altered the carbon and energy cycle in the Earth
868 system, leading to the new epoch: the “Anthropocene”.

869 Many kinds of man-made organics such as synthetic polymers were produced and
870 delivered into the environment. One of the most ubiquitous polymer is debris of plastics,
871 which was produced in large quantities after World War II (Carpenter & Smith, 1972).
872 Jambeck et al. (2015) reported that 275 million metric tons of plastic waste was generated in
873 2010 and 5 ~ 13 million tonnes of plastic have been transported to the ocean. Plastic

874 fragments are stable and highly durable, potentially lasting hundreds to thousands of years
875 (Barnes et al., 2009; Cózar et al., 2014). Thus, like the emergence of lignin with the
876 appearance of vascular plants on land (~420 Mya; Lenton, 2001; Banks et al., 2011) or
877 pyrogenic carbon (420 Mya; Cressler, 2001; Bird et al., 2015) the emergence of plastics
878 marks the beginning of a new era in the evolution of organics on Earth (Wu et al., 2017,
879 2019a, b).

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882 Figure legends

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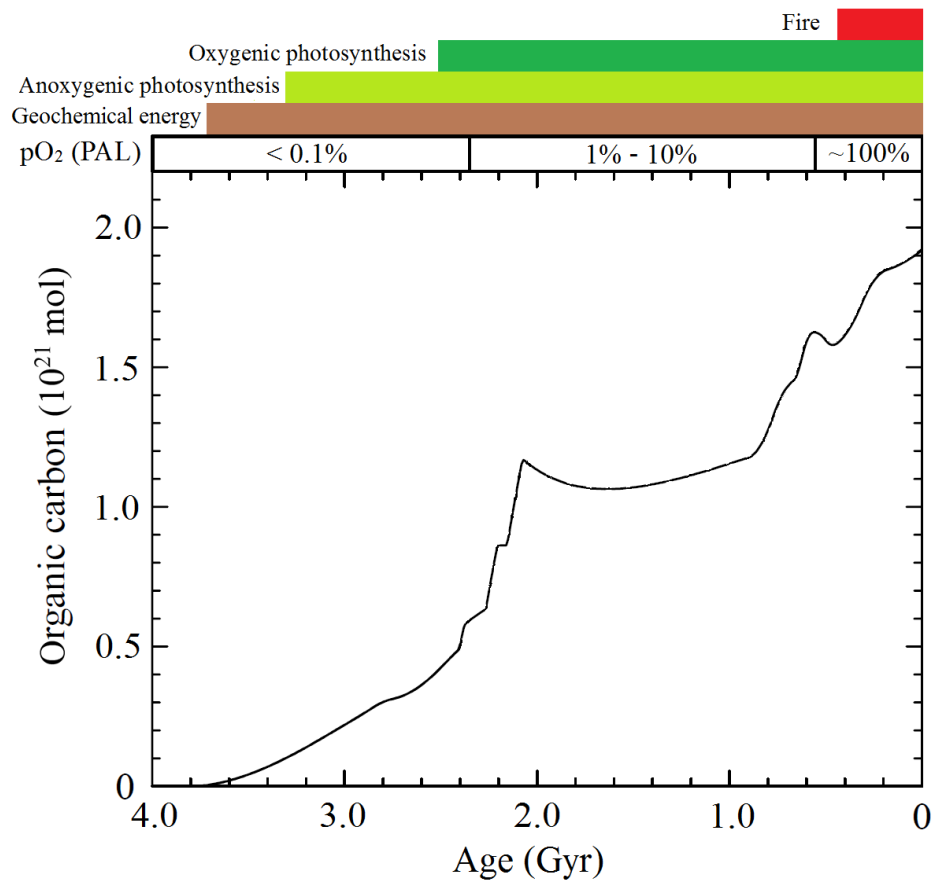
884 Fig. 1 Quantity of organic carbon in the crust against age according to references (Des Marais
885 et al., 1992; Hayes & Waldbauer, 2006). PAL: present atmospheric level. The variety of
886 energy sources, e.g. geochemical energy, sunlight, oxygen and fire, to support the evolution
887 of life.

888 Fig. 2 The derivation and characteristics of organic matter from asteroids, microbes, plants,
889 fires and humans.

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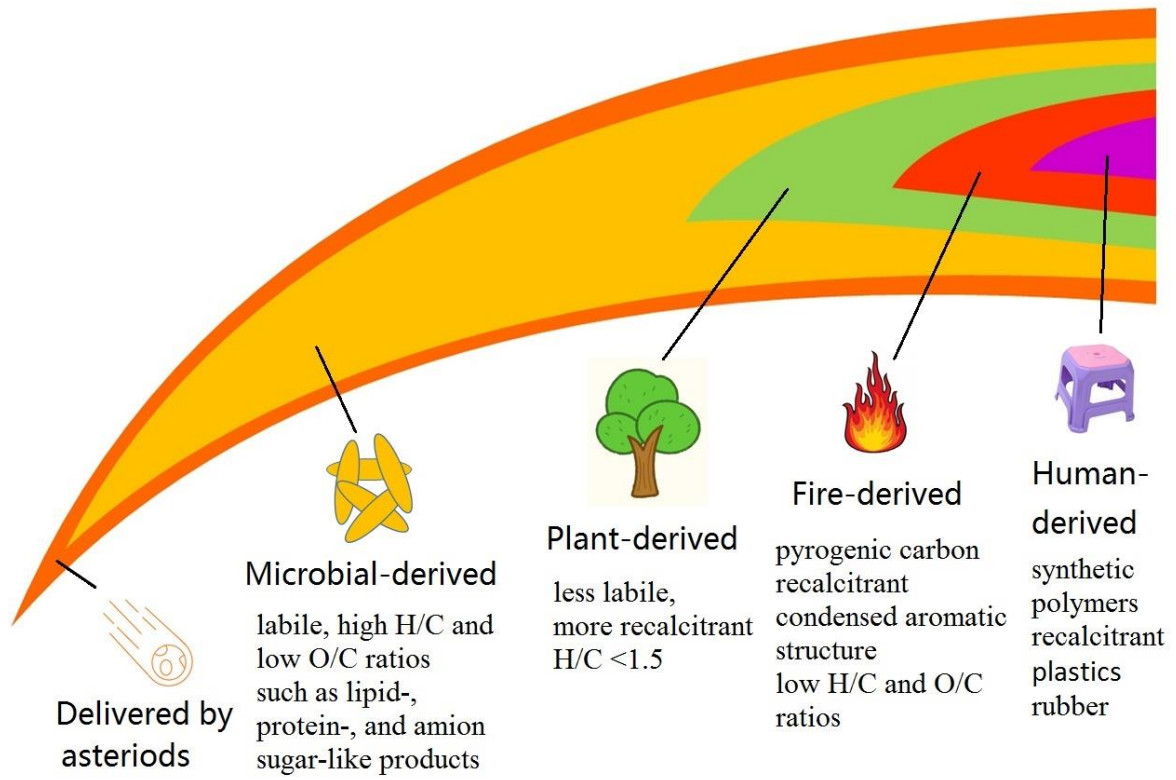


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Figure 1

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Figure 2

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