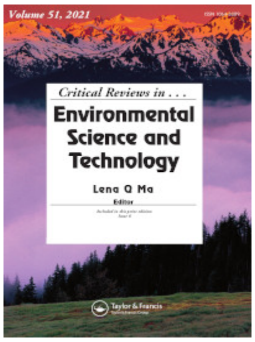

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The co-evolution of life and organics on earth: Expansions of energy harnessing

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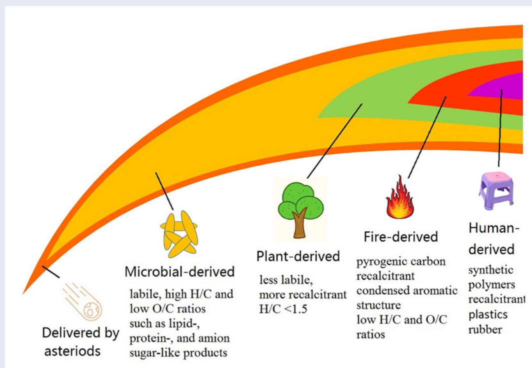
ABSTRACT

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



KEYWORDS Energy harnessing; life; organic matter

1. Introduction

Life is processes of generating reduced organic compounds from carbon dioxide as well as the harnessing of environmental energy. Over the course

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of Earth history, the harnessing of free energy by organisms has had a dramatic impact on the geosphere, including minerals and organics (Dietrich, Tice, & Newman, 2006; Grosch & Hazen, 2015; Judson, 2017), shaped the whole trajectory of life evolution. As a direct consequence of a coevolving geosphere and biosphere, the Earth's crust has changed greatly over billions of years. The origin and evolution of organic compounds on the planetary environment are compelling because of their potential role in the origin of life and sustaining microbial communities (Lazcano & Miller, 1996; McDermott, Seewald, German, & Sylva, 2015; Schönheit, Buckel, & Martin, 2016). Carbon lies at the heart of carbon-based life forms and provides unparalleled potential for earth evolution. The origin of life is inextricably linked to the behavior of carbon (Hazen, 2019). The evolution of organics is coupled with the evolution of life, which is expanded with a variety of available free energy sources (Judson, 2017). Collectively, these linkages have mediated the generation and transformation of soils and sediments. Here we review the origin and evolution of organics on Earth, and their relationship with diversification and expansion of energy utilization and with biological and geological development.

2. The prebiotic organics

Earth accreted 4.56 billion years (Gyr) ago from largely homogeneous material (Hazen et al., 2013; Judson, 2017). With the dissipation of thermal energy produced by compaction, radiation, and impacting meteorites, the Earth cooled. About 4.4 Gyr ago patches of a rocky scum had solidified and eventually separated into core, mantle, and crust (Mojzsis, 2010). Water vapor condensed as rain and formed early oceans and seas (Hazen et al., 2013; Rosing, Bird, Sleep, Glassley, & Albarede, 2006; Wilde, Valley, Peck, & Graham, 2001). The early ocean was a reservoir of inorganic elements, and also a reservoir of potential free energy in the form of protons. Before the emergence of life on early Earth at ~3.8 Gyr (Dodd et al., 2017), these prebiotic organics were either synthesized abiotically on the Earth itself or synthesized extraterrestrially and then delivered to the Earth (Hayes, 1967; Dalai, Kaddour, & Sahai, 2016). A wide range of organic compounds including amino acids, monocarboxylic acids, sugars, nucleobases, and membrane-forming lipids have been synthesized in prebiotic conditions simulation experiments (McCollom, 2013). Questions remain, however, concerning whether the conditions that allow synthesis of these compounds in the laboratory accurately simulate those that might have been present on the early Earth (Dalai et al., 2016; McCollom, 2013). High concentrations of the reactants, water pH and ambient temperature are of central importance in experimental abiotic synthesis of organics. The

Box 1. Abiotic synthesis of organic compounds in hydrothermal environments.

Since the discovery of deep-sea hydrothermal systems in the late 1970s (Corliss et al., 1979), there has been keen interest in the origin of life in these environments. The concept that deep-sea hydrothermal systems as sites of abiotic organic synthesis is based largely on their strongly reducing chemical environments. Based on geological observations as well as theoretical and experimental constraints, the theory received support as having the potential for abiotic synthesis of organic compounds within hydrothermal environments (Charlou et al., 2002; Martin, Baross, Kelley, & Russell, 2008). The abiotic formation of organic compounds in geologic systems involves the abiotic reduction of dissolved inorganic carbon ($\sum CO_2 = CO_2 + HCO_3$\bar$+ CO_3{}^{2-}$) to organic compounds by dissolved H_2 produced by serpentinization, which can be expressed by the following reaction (McCollom & Seewald, 2007):

$$CO_2 + H_2 \rightarrow CH_4 + C_2H_6 + C_3H_8 + C_nH_n + 2 \ldots + H_2O$$

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

extreme environments (highly acidic condition) of early Earth presented severe limitations with respect to their potential for prebiotic chemistry because of stability and synthetic pathway issues associated with temperatures and pH (Bada, 2013). It has been claimed that autocatalytic metabolic-like reactions can overcome these limitations (Huber & Wächtershäuser, 1998). The micro-conditions in the hydrothermal systems supposedly could drive abiotic syntheses of organics (McCollom & Seewald, 2007; McDermott et al., 2015). Prebiotic syntheses could have taken place in a variety of geochemical environments that may have existed on the primitive Earth, although this has never been demonstrated using plausible geochemical conditions (Box 1). Highly reducing fluids such as deep-sea hydrothermal fluids have the potential for abiotic reduction of dissolved inorganic carbon to produce organic compounds (McDermott et al., 2015; Seewald, Zolotov, & McCollom, 2006; Shock, 1990; Shock & Schulte, 1998). There is also increasing evidence that supports an abiotic origin for CH_4 and other low-molecular weight reduced organic compounds in ultramafic-hosted hydrothermal systems (Charlou, Donval, Fouquet, Jean-Baptiste, & Holm, 2002; McCollom & Seewald, 2007; Proskurowski et al., 2008). Given the scarcity of suitable abiotic regime the yield prebiotic organics on the early Earth would have been very small (Lollar, Westgate, Ward, Slater, & Lacrampe-Couloume, 2002).

Besides the abiotic synthesis of organic molecules on the young Earth driven by various energy sources such as UV radiation in sunlight, cosmic rays, X-rays, hypervelocity impacts, volcanic eruptions with lightning, geothermal heat, and redox gradients, the total inventory of organics would have included exogenous sources (the interstellar medium, interplanetary dust, asteroids, comets, meteorites) (Dalai et al., 2016; Kwok, 2016; Sahai, Kaddour, & Dalai, 2016; Sandford, Engrand, & Rotundi, 2016). It has long been speculated that Earth accreted prebiotic organic molecules from impacts of carbonaceous asteroids and comets during the period of 4.5 Gyr to 3.8 Gyr ago (Botta & Bada, 2002; Chyba, Thomas, Brookshaw, & Sagan, 1990; Chyba & Sagan, 1992) because the exogenous delivery has showered the Earth (Pizzarello & Weber, 2004). Polyhydroxylated compounds (such as sugars, sugar alcohols and sugar acids) are formed under interstellar conditions via photolysis of small molecules (e.g. CO, NH_3 and H_2O) and are therefore present in meteorites (Agarwal et al., 1985; Cooper et al., 2001; McDonald et al., 1996). The carbonaceous component of interplanetary dust could be up to 50 wt% (Dalai et al., 2016; Ehrenfreund & Charnley, 2000). This dust material has been reported to contain simple aliphatic, aromatic compounds, macromolecular polyaromatic hydrocarbons (Dalai et al., 2016; Ehrenfreund & Charnley, 2000), amino acids and other organic compounds (Cooper et al., 2001) that are vital to the origin of life. Tens of thousands of tons of interplanetary dust particles enter the Earth's atmosphere annually, and the rate may have been much greater on early Earth (Kwok, 2016). It was estimated that Earth was also accreting intact cometary organics at a rate of at least $\sim 10^9$ to 10^{10} g per year at 4.5 Gyr (Chyba et al., 1990). Organics delivered from space comprising as much as perhaps 10% of the Earth's modern biomass by weight (Schönheit et al., 2016; Sephton, 2002) estimated that about 1.0×10^{21} mol of reduced carbon were probably delivered to the surface of Earth by asteroids ($4.4 - 3.8$ Gyr) (Catling, Zahnle, & McKay, 2001; Hayes & Waldbauer, 2006).

3. Geochemical energy

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

At this time in Earth history, oxygen was at trace levels (Canfield, Rosing, & Bjerrum, 2006), so the first ecosystems must have existed in an anoxic world and their activities were driven by anaerobic metabolisms (Canfield et al., 2006; Judson, 2017). The proposed emergency of life under anoxic geothermal environments implies that life started not as a planetary but as a local phenomenon. It was reported that metabolisms of early anaerobic ecosystems were probably 2–3 orders of magnitude less active than the present biosphere (Des Marais, 2000). Given these factors and the probable limits on accessing the most limiting chemical compounds, various ecosystems most likely existed in relative isolation (Canfield et al., 2006).

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

4. Sunlight

The greatest energy source in the surface environment of the Earth is sunlight. Today the average solar energy flux to Earth surface is 340 W/m^2 (Rosing et al., 2006). The early Sun was fainter and solar luminosity was probably a quarter to a third less than the present day (ca 250 W/m^2 at 4.0 Gyr; Nisbet & Sleep, 2001; Sagan & Chyba, 1997).

It is reported that by ca. 3.7 Gyr (Figure 1) (Nutman, Bennett, Friend, Van Kranendonk, & Chivas, 2016; Pecoits et al., 2015; Rosing, 1999), photosynthetic organisms emerged to harness the energy in sunlight to drive chemical reactions. When the biosphere developed photosynthesis, living organisms acquired the ability to absorb solar energy and convert a fraction of it into chemical free energy (Rosing et al., 2006). Photoautotrophs acquired the ability to build up gradients in chemical potential, rather than just exploiting existing gradients, as was the fate of their chemoautotrophic predecessors. With evolution of photosynthesis, energy resources available

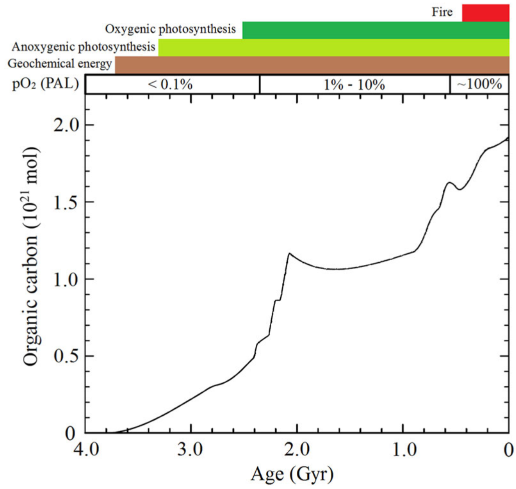

Figure 1. Quantity of organic carbon in the crust against age according to references (Des Marais et al., 1992; Hayes & Waldbauer, 2006). PAL: present atmospheric level. The variety of energy sources, e.g. geochemical energy, sunlight, oxygen and fire, to support the evolution of life.

for lives became several orders of magnitude larger than that available from oxidation-reduction reactions (chemoautotrophic primary production) associated with hydrothermal activities (Des Marais, 2000; Rosing, 2005; Rosing et al., 2006). The development of photosynthesis allowed life to escape the hydrothermal setting (Nisbet & Sleep, 2001). Energy harvested from sunlight, therefore, enhanced the rates of autotrophic carbon fixation, and carbon burial in anoxic environments. The primary productivity of the photosynthetic world was estimated to be 10,000 times higher than those of non-photosynthetic ecosystems (Sleep & Bird, 2008; Summons & Hallman, 2014), although the rates would have been significantly lower than the present (Summons & Hallman, 2014). The earliest photosynthetic organisms performed anoxygenic photosynthesis, and were dependent on mineral sources as electron donors, but relieved the energy constraints to perform reduction of organic compounds (Olson & Blankenship, 2004; Rosing et al., 2006). The genesis of photosynthesis had irreversible consequences for Earth surface environments whether it was oxygenic or anoxygenic.

Estimates suggest that anoxygenic phototrophs increased the flux of carbon through the biosphere and the most active ecosystems were probably driven by the cycling of Fe^{2+}, with the oxidation of Fe^{2+} yielding potentially the highest rates of primary production (Canfield et al., 2006). The importance of hydrogen as an early fuel for anoxygenic photosynthesis has also been emphasized by Olson (2006), and may have been sufficiently abundant in the early Earth to drive CO_2 reduction. Other dynamic ecosystems would have also been driven by the microbial cycling of sulfur and nitrogen species, but these would have been considerably less active in

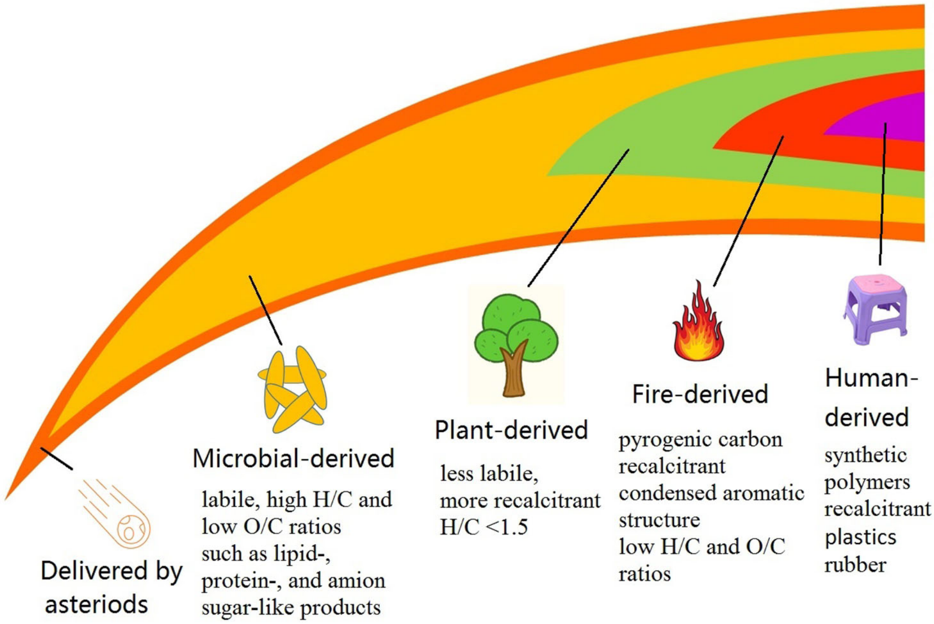

Figure 2. The derivation and characteristics of organic matter from asteriods, microbes, plants, fires and humans.

comparison with those based on iron and hydrogen as electron donors to reduce CO_2 (Canfield et al., 2006). For all the ecosystems mentioned above, the production rates of organics were considerably less than those of today. The primary production rates of total carbon at 3.8 Gyr ago were estimated as 2.8×10^{14} mol yr^{-1}. Organic carbon accounted for 14% of the total carbon. This primary production estimate is 14 times lower than present rates (4.0×10^{15} mol yr^{-1}) (Canfield et al., 2006). Considering that prokaryotic life was flourishing and presumably widespread in the biosphere, organics must have been completely microbially derived, which produces more labile organic matter with high H/C (the degree of aliphatic character) and low O/C or (O + N)/C (the degree of polar character) ratios (Qiu et al., 2015) such as lipid-, protein-, and amion sugar-like products (Figure 2) (Brocks, Logan, Buick, & Summons, 1999; D'Andrilli, Cooper, Foreman, & Marshall, 2015; Grannas, Hockaday, Hatcher, Thompson, & Mosley-Thompson, 2006).

5. Oxygen

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

The stratospheric ozone layer seems to have been created at 2.3 Gyr (Goldblatt, Lenton, & Watson, 2006). The formation of the ozone layer was facilitated by O_2 levels rising to 1-3% of present levels; at these levels, photolysis of oxygen (yielding reactive oxygen radicals) was sufficiently frequent for an ozone layer to be produced, which shielding life from short-wave UV-radiation (200-300 nm) and was suitable for life to expand on the continents (Goldblatt et al., 2006). Beyond the energetic limitations, defined by the availability of oxygen, the only limiting factors for life from the environment became the availability of bio-essential elements (Rosing et al., 2006). The increase in the oxygen content of the atmosphere and ocean driven by photosynthesis increased chemical weathering rates (Shields, 2007), which in turn increased nutrient (e.g. phosphorus, partly nitrogen, and iron) availability (Kump, 2010; Shields, 2007). The far-reaching impact of the GOE cannot be emphasized enough: it changed Earth's history by enabling the evolution of aerobic life, an explosion in the biosynthesis of organics and this underpinned the opportunity for organics to be generated and to proliferate in large quantities on a planetary scale.

Oxygenic photosynthesis is by far the most efficient mechanism for harvesting solar energy (Rosing et al., 2006). The GOE changed Earth's history by enabling the evolution of aerobic life (eukaryotes) and the emergence of the lineage that would eventually produce land plants. On Earth, aerobic metabolism provides about an order of magnitude more energy for a given intake of food than anaerobic metabolism (Judson, 2017). As a consequence of energetic limitations, life without O_2 as a strong electron acceptor, well mixed in the atmosphere and the surface ocean, could not grow large and complex (Lane & Martin, 2010). On account of prohibitively low growth efficiencies and energetic limitations anaerobes do not grow beyond the complexity of uniseriate filaments of cells (Catling & Claire, 2005; Schulz & Jørgensen, 2001). The oxygenated atmosphere and ocean enabled the evolution of more complex life (Dahl et al., 2010; Kump, 2010; Payne et al., 2009). The maximum body size of organism has increased by 16 orders of magnitude since emergence of life; this transition occurred via two discrete steps (Payne et al., 2009). The first was the emergence of eukaryotic cell (~1.9 Gyr) and the second was eukaryotic multicellularity (0.6-0.45 Gyr). These two steps coincide or slightly postdate with increases in atmospheric oxygen levels (Payne et al., 2009). The evolution of Earth's biota is intimately linked to the oxygenation of the atmosphere and the oceans (Dahl et al., 2010). This atmospheric oxygenation correlates with the diversification and radiation of vascular plants on the continents (Gensel, 2008) and the oxygenation of the oceans correlates with the expansion of large predatory fish (Bambach, 2002). This evolution significantly enhanced the burial of reduced carbon and was companied by the accumulation of organic matter (Figure 1).

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

The emergence of larger, and less easily degradable organic molecules related to eukaryotic diversification thus enhanced the burial of organic matter and its diversity. One of the most unique and pervasive biological characteristics of organic matter in terrestrial environments is the predominance of sources from vascular plants (Oades, 1993). For example, during the Carboniferous period (360-300 Ma), oxygen in atmosphere rose to between 30 and 35% (Berner, Beerling, Dudley, Robinson, & Wildman, 2003; Hsia, Schmitz, Lambertz, Perry, & Maina, 2013), coinciding with the appearance giant vascular plants, fern-dominated forests (Shear, 1991). Fern are lignin rich plants that contain $>$ 40% of lignin than modern plants (~20%) (Robinson, 1990), and this lignin was difficult to decompose until organisms like fungi evolved and effective degradation occurred until 200 million years after fern plant emergence (Robinson, 1990). The rise of ligniferous plants and low lignin breakdown (due to the rare or absence of lignolytic organisms) contributed to increased terrestrially derived organic matter burial through inhibited decomposition (Berner et al., 2003; Robinson, 1990). The spread of vascular plants in the terrestrial environment increased the diversity of organic matter, including plant-derived polysaccharides such as cellulose and phenolic compounds such as lignin (Benner, Maccubbin, & Hodson, 1984; McLatchey & Reddy, 1998). These organic compounds are characterized by less labile, more recalcitrant chemical nature with $H/C < 1.5$ (Figure 2) (D'Andrilli et al., 2015).

6. Fire

To trigger wildfire, all of three conditions must be met (Scott & Glasspool, 2006). Firstly, fire ignition was required—such as lightning strikes, meteor strikes and volcanic activity. Throughout the Earth history, these have been abundant. Lightning is the preeminent source of heat for the ignition of fossil wildfires. Lightning strikes occurred more than 1.4 billion lightning strikes per year owing to its global frequency (44 ± 5 strikes/s) (Christian et al., 2003), of which an appreciable number ignite wildfires. Secondly, sufficient amount of oxygen became present in the atmosphere. Assuming current atmospheric pressure, at least 16% oxygen is the minimum concentration in order for plants to ignite and for fire to be self-sustaining (Belcher, Yearsley, Hadden, McElwain, & Rein, 2010; Belcher & McElwain, 2008). For most of Earth's history, oxygen levels have been lower than this threshold until 0.35 Gyr (Scott, 2000; Scott, Bowman, Bond, Pyne, & Alexander, 2013). Thirdly, fire requires fuel. The earliest land plants (embryophytes) evolved from charophycean green algal ancestors (Steemans et al., 2009) at approximately 0.47 Gyr (Berner, 2009). The appearance of vascular plants on land occurred around 0.42 Gyr ago, although they were tiny and leafless (Banks et al., 2011; Lenton, 2001). All three conditions were met and fire activity has begun to influence the Earth system and the cycling of organic matter.

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

Fire has had both geological and biological impacts on ecosystems. Fire regimes drive the evolution of plant traits, such as thick bark (Bond, Woodward, & Midgley, 2005; Keeley, Pausas, Rundel, Bond, & Bradstock, 2011; Pausas & Keeley, 2009); the initial spread of flowering plants (Bond & Scott, 2010); faunal abundance and diversity such as ants (Moreau, Bell, Vila, Archibald, & Pierce, 2006); shape biomes (Crisp, Isagi, Kato, Cook, & Bowman, 2010, Crisp, Burrows, Cook, Thornhill, & Bowman, 2011; Scheiter et al., 2012); affect soils quality and nutrient cycling such as the carbon, oxygen and phosphorous cycles; promote biodiversity (Bond & Keeley, 2005). Due to the fire integration to ecosystem function and maintenance (Edwards et al., 2010; Keeley & Rundel, 2005), pyrophilic grasslands and savannas such as C_4 grasslands expanded and replaced woodlands (Hoetzel, Dupont, Schefuß, Rommerskirchen, & Wefer, 2013; Keeley & Rundel, 2005).

The emergence of fire in terrestrial environment likely had a profound effect upon the compositions and dynamics of organic carbon. Furthermore, fire contributes new material to the Earth—pyrogenic carbon or fire-derived organic matter (partly charred organic matter including black carbon, charcoal and soot) (Judson, 2017; Lenton, 2013). Glinka (1914) described that “there was almost no soil profile in which charcoal particles did not occur in the upper horizon” (Bird, Wynn, Saiz, Wurster, & McBeath, 2015). In modern peats, charcoal may constitute 4% of the total volume. In the Carboniferous, charcoal represented more than 20% of dead organic matter (Scott et al., 2013). It was recently estimated that 3-5 million km^2 of the Earth surface are burned by wildfires annually (Jones, Santin, van der Werf, & Doerr, 2019) and approximately 116-385 Tg/yr of pyrogenic carbon are now produced globally by fires (Santin et al., 2016). Pyrogenic carbon can represent a significant proportion of total organic carbon in the environment: ranging from 2% to 60% of the total soil organic carbon in terrestrial systems (Reisser, Purves, Schmidt, & Abiven, 2016; Singh, Abiven, Torn, & Schmidt, 2012). Santin et al. (2016) provided a global assessment of pyrogenic carbon fluxes. Accounted for 8 to 27% of the annual production of pyrogenic carbon were inputted to oceans from rivers and most of pyrogenic carbon was deposited on the continental shelf (Santin et al., 2016). These reports indicate that pyrogenic carbon is a significant component in both terrestrial and oceanic carbon storage (Preston & Schmidt, 2006). Moreover, pyrogenic carbon has a condensed aromatic structure with low H/C and O/C ratios; it is therefore recalcitrant (D’Andrilli et al., 2015). It has been established that pyrogenic carbon can persist in soils and sediments for millions of years and thus it plays an

Box 2. **Fire and hominins.**

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

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

Many kinds of man-made organics such as synthetic polymers were produced and delivered into the environment. One of the most ubiquitous polymer is debris of plastics, which was produced in large quantities after World War II (Carpenter & Smith, 1972). Jambeck et al. (2015) reported that 275 million metric tons of plastic waste was generated in 2010 and 5 ∼ 13 million tonnes of plastic have been transported to the ocean. Plastic fragments are stable and highly durable, potentially lasting hundreds to thousands of years (Barnes, Galgani, Thompson, & Barlaz, 2009; Cózar et al., 2014). Thus, like the emergence of lignin with the appearance of vascular plants on land (∼420 Mya; Lenton, 2001; Banks et al., 2011) or pyrogenic carbon (420 Mya; Cressler, 2001; Bird et al., 2015) the emergence of plastics marks the beginning of a new era in the evolution of organics on Earth (Wu et al., 2017, 2019, 2020).

important role in the global carbon inventory of the Earth and fluxes between reservoirs over long time scales (Forbes, Raison, & Skjemstad, 2006; Schmidt & Noack, 2000; Scott, 2010). Human activities that suppress the production of pyrogenic carbon have significantly disturbed the pyrogenic carbon cycle (Andela et al., 2017; Bowman et al., 2011).

The advent of anthropogenic fire was a revolutionary event in Earth history because fire technology has significantly influenced the biosphere over the last 10,000 years (Box 2) (Raupach & Canadell, 2010). Fire gave protection, extended the range of food, and expanded adaptation to different environments on Earth (Froestad & Shearing, 2017). Fire plays a pivotal role in the clearing of forests to create permanent fields with the development of sedentary agriculture-based societies during the Holocene (Bowman, O'Brien, & Goldammer, 2013). During the late Quaternary humans have dramatically altered fire regimes around the globe, which is largely dependent on fossil fuels, both directly and indirectly. The production and existence of pyrogenic carbon underpin the significant perturbations of the carbon cycles both, on long (million year) (Berner, 1999a, 1999b, 2003) and on short (thousand year) timescales. The application of fire by humans, especially the fossil fuel burning, has accelerated both long- and short-term carbon cycles through anthropogenical alternation of carbon fluxes, the increase of CO_2 in the atmosphere, and global warming (Berner, 1999a, 1999b). Organic aerosols such as soot from fire smoke in

Earth's atmosphere is an important contributor to global climate change by absorbing heat and warming the air (Berner, 2003; Bond et al., 2013; Johansson, Head-Gordon, Schrader, Wilson, & Michelsen, 2018).

Life is a process of harnessing energy to maintain states far from thermodynamic equilibrium, leading to an energy flow through the biosphere (Raupach & Canadell, 2010). Any reduction of an energy source could cause a corresponding contraction in the biosphere and drop in the rate of global organic matter burial. In the traditional "big five" mass extinctions oceanic anoxic events (due to the worldwide reduction of oxygen) have coincided with four of these mass extinctions, especially, the end-Ordovician (Zhang, Shen, Zhan, Shen, & Chen, 2009), the Late Devonian (Goddéris & Joachimski, 2004), the end-Permian (Grice et al., 2005; Wignall & Twitchett, 1996), and the end-Triassic (Isozaki, 1997). Thus, rapid declines in atmospheric O_2 have been proposed to have a major influence upon mass extinction events. For example, in the most severe extinction (loss of as much as 95% of all species on Earth) that occurred in the Late Permian (~251 Mya) (Benton & Twitchett, 2003; Chen & Benton, 2012; Grice et al., 2005), the oxygen level (at 30% or more in the Carboniferous period) fell dramatically to 13% in the late Permian and the early part of the subsequent Triassic (Lane, 2007). One factor in the last mass extinction (end-Cretaceous) may be the prevention of sunlight from reaching the surface of the Earth due to the dust, soot or aerosols in the atmosphere ejected by the Chicxulub asteroid impact (Kring, 2007).

The sudden mass mortality of the terrestrial and marine organisms irreversibly reorganized the global carbon cycle (Berner, 2002; Caplan & Bustin, 1999). A drop in burial fluxes for global organic carbon is coincident with the abrupt biota change at the time of late Permian (Berner, 2005) and the end-Cretaceous mass extinctions (D'Hondt, Donaghay, Zachos, Luttenberg, & Lindinger, 1998). Such a reduction in the organic flux could have been a natural consequence of the ecosystem reorganization that resulted from the mass extinctions.

7. Conclusions

The amount and proportion of pyrogenic carbon in total organic carbon on Earth are expected to increase, which changed the diversity and complexity of organic matter on Earth. The change of the diversity and complexity of organic matter (microbial-, plant-, fire- and human-derived organics) have in-return significantly influenced Earth's carbon cycle, planetary climate and ecosystems. The long-term organic carbon cycle has dominant influenced the levels of atmospheric oxygen and carbon dioxide over a multimillion-year time scale. The levels of atmospheric carbon dioxide and oxygen were mainly mediated via weathering of organic matter on the continents, the burial of organic matter in sediments, and the thermal breakdown of organic matter at depth. The increased oxidation of organic carbon to carbon dioxide by weathering and thermal breakdown results in the O_2 consumption and CO_2 production. The burial of organic matter in sediments leads to an increase in atmospheric O_2 and a decrease of atmospheric CO_2 due to a net excess of photosynthesis over respiration (Berner, 1999a, 1999b). Current burning of fossil carbon results in a decrease of atmospheric O_2 by about 2 ppm per year (Keeling & Manning, 2014). The fluctuation of atmospheric levels of O_2 and CO_2 would change the planetary climate, temperature, precipitation (enhanced atmospheric CO_2 leads to a warmer and wetter climate via the greenhouse effect), resulting in the evolution of biology. An integrated view of assessing the role of the change of diversity and complexity of organic matter is required. A full understanding of the role of pyrogenic carbon on planetary climate and ecosystems can provide us with new opportunities for mitigating climate change.

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

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