




Frontiers in Prebiotic Chemistry and Early Earth Environments

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Received: 4 February 2022 / Accepted: 29 April 2022
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Abstract

The Prebiotic Chemistry and Early Earth Environments (PCE₃) Consortium is a community of researchers seeking to understand the origins of life on Earth and in the universe. PCE₃ is one of five Research Coordination Networks (RCNs) within NASA's Astrobiology Program. Here we report on the inaugural PCE₃ workshop, intended to cross-pollinate, transfer information, promote cooperation, break down disciplinary barriers, identify new directions, and foster collaborations. This workshop, entitled, “*Building a New Foundation*”, was designed to propagate current knowledge, identify possibilities for multidisciplinary collaboration, and ultimately define paths for future collaborations. Presentations addressed the likely conditions on early Earth in ways that could be incorporated into prebiotic chemistry experiments and conceptual models to improve their plausibility and accuracy. Additionally, the discussions that followed among workshop participants helped to identify within each subdiscipline particularly impactful new research directions. At its core, the foundational knowledge base presented in this workshop should underpin future workshops and enable collaborations that bridge the many disciplines that are part of PCE₃.

Keywords Origins of Life · Prebiotic Chemistry · Earth environments · Atmospheric Chemistry

Introduction to PCE₃

The Prebiotic Chemistry and Early Earth Environments (PCE₃) Consortium seeks to understand the origins of life on Earth and in the universe. PCE₃ is one of five Research Coordination Networks (RCNs) within NASA's Astrobiology Program. PCE₃ advances the science of the origins of life by establishing connections across diverse disciplines, stressing the context of early Earth conditions. Researchers within this network investigate the delivery

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(Schmitt-Kopplin et al. 2010; Callahan et al. 2011; Herd et al. 2011), synthesis (Barge et al. 2019; Yadav et al. 2020), and fate of small molecules, and the formation of proto-biological molecules (Frenkel-Pinter et al. 2019; Cakmak et al. 2020) and pathways that led to systems harboring the potential for life (Damer and Deamer 2020). This community is interested in key transitions of the geosphere (Foley 2019; Tusch et al. 2021), atmosphere (Zahnle et al. 2020), and hydrosphere (Sleep et al. 2001; Korenaga 2021) during the planetary transition from abiotic to biotic.

Vast disciplinary breadth falls under the PCE₃ umbrella, ranging from astrophysics and geoscience, to synthetic organic chemistry, biochemistry, and evolution. Study of life's emergence is concerned with organic and inorganic chemical processes, networks of chemical reactions, and chemical evolution, all in the context of planetary environments, which varied over time and space. PCE₃ aims to identify planetary conditions that can or cannot give rise to biochemistry, placing boundary conditions on the environments of life's beginnings on Earth and guiding future missions that target the discovery of other habitable worlds. PCE₃ aims to explore the biochemical limits of life and help create experimental models of extinct proto-life. By fostering cross-disciplinary communication and collaboration, PCE₃ will enable the community to establish models for the emergence of prebiotic and early biotic chemistry that are rooted in realistic planetary scenarios, integrating the conditions, dynamics, possibilities, and uncertainties of early Earth environments into origins hypotheses.

To achieve these objectives, PCE₃ seeks to identify novel multidisciplinary research approaches, new paradigms, and revolutionary advances. PCE₃ is advocating for community-wide and programmatic conversations about these advances with the hope of spawning a new wave of broadly integrated research. PCE₃ is bringing together diverse communities and providing mechanisms for transfer of information among them. In doing so, the aim is to promote and facilitate scientific cooperation and to break down structural and philosophical barriers while developing and supporting innovative methods of scientific communication and interaction. Top priorities include mentoring and promoting young scientists and providing gateways into the established community with an overarching goal of increasing diversity, equity, and inclusion.

In summary, the Consortium will:

- *Integrate the early Earth and prebiotic chemistry communities and break down disciplinary barriers in pursuit of plausible prebiotic chemistry pathways.*
- *Develop robust and fully-parameterized models of early Earth environments that can be explored both experimentally and theoretically for their potential to host prebiotic chemical pathways.*
- *Promote novel and innovative experimental and theoretical approaches to exploring the abiotic → prebiotic → biotic transition.*
- *Identify planetary conditions that can or cannot give rise to life's chemistry and thus inform the exploration for life throughout the universe.*
- *Characterize geochemical and geophysical constraints of early Earth environments that can be applied to test, verify, validate, and guide existing and future experimental and theoretical prebiotic chemistries.*

Background and Motivation of PCE₃ Workshops

The first objective serves as the foundation for the others and therefore is the central focus of early PCE₃ activities, specifically the interdisciplinary workshop described here. PCE₃ has designed a series of workshops to cross-pollinate, transfer information, promote cooperation, break down disciplinary barriers, identify new directions, and foster collaborations. The inaugural PCE₃ workshop is composed of two parts. *Part I: Building a New Foundation* was held from October 1 to November 20, 2021, and was designed to broadly disseminate current knowledge, identify possibilities for multidisciplinary collaboration, and ultimately define paths for future collaborations. The next workshop, *Part II: New Directions*, being organized for 2022, is intended to finalize cross-pollination, facilitate formation of new multidisciplinary teams, and promote novel research avenues and specific projects. These workshops encourage and facilitate participant interactions to help reach these objectives.

Part I: Building a New Foundation was focused on five broad themes, (1) Earliest Planetary Formation; (2) Evolution of the Near Surface; (3) Inventories, Geological Settings, and Building Blocks; (4) Prebiotic Complexity; and (5) Peering into the Past with Today's Biochemistry. Each of these themes was broken down into specific subsections (a summary of each is given in Section C).

Speakers of the Part I workshop provided overviews of specific topics at levels appropriate for broad audiences, consistent with PCE₃'s aim to improve cross-disciplinary communication. The presentations underscored points of agreement, as well as debates and uncertainties, while highlighting important next steps within each subtheme. These “primer talks” were designed to educate, clarify assumptions, and correct misconceptions within a framework of agnostic disciplinary overviews—presenting a breadth of scientific viewpoints and untethered from specific models of life's origins.

With the aim of stimulating cross-pollination between disciplinary experts and extra-disciplinary “consumers”, presentations addressing the likely conditions on early Earth were designed in ways that could be incorporated into prebiotic chemistry experiments to improve their planetary plausibility. Additionally, the discussions that followed among workshop participants helped to identify, within each subdiscipline, research that might catalyze important new directions across the origins of life community. At its core, the foundational knowledge presented in this workshop should underpin future workshops and enable collaborations that bridge the many disciplines that are part of PCE₃. In the second installment of this workshop series, “*Part II: New Directions*,” we anticipate that these efforts will manifest in new multidisciplinary collaborative teams and transformative research avenues, thus directly contributing to PCE₃'s Measure of Success (<http://prebioticchem.info/about/mos.html>).

Workshop Logistics. Two approaches of Workshop I were (1) primer talks providing access to information across a breath of disciplines and (2) participant discussions and interactions for discovering innovative research directions. Consistent with COVID-19 restraints, Workshop I was virtual. The workshop was configured with both synchronous and asynchronous sessions of five themes distributed across five weeks (see section C). Primer talks were pre-recorded, and workshop participants were asked to view these in advance of synchronous weekly programs. The synchronous agenda began with a short (~20 min) plenary session that included a brief introduction and 2-minute overviews of primer talks. The remainder of the weekly synchronous meetings was dedicated to small

(8–12 participant) breakout groups. The moderated breakout groups were charged with identifying critical questions and research avenues within each weekly theme that remain unresolved or underexplored and could have high impact on origins research. These discussions were chronicled and served as a partial basis for this report. Workshop logistics were carried out with the help of *Know Innovation* (knowinnovation.com), an organization that facilitates creativity and innovation in interdisciplinary scientific groups.

A key goal of this workshop is broad community participation across backgrounds, disciplines and career stages. By these metrics (Supplementary Materials), the workshop was a success. Over 500 people with diverse backgrounds participated in the workshop in real time. Many more have watched or are watching the online content.

Summaries of the Seminars Given in the PCE₃ Workshop on Oct 9 to Nov 20, 2020

Theme 1: Earliest Planetary Formation (Oct. 9, 2020).

Theme 1 discussed events and processes during the early formation of Earth that are relevant to origins of life. Important contributions included the influence of the Sun on Earth's conditions; the formation of the Earth and its metallic core; how the Moon formed and influenced conditions on Earth; and how early Earth was modified by its internal dynamics, tectonics, and meteoritic bombardment. What follows are brief summaries of each talk provided by the speakers. Recordings of these talks can be found here (https://www.youtube.com/playlist?list=PLvogKQh-bBnUwhFNuvvgqIM_-Xb01jDqFO).

1a. Stellar Evolution (Edward Schwieterman, University of California, Riverside): The Sun's overall luminosity and activity has changed over geologic time, influencing the environmental conditions at the origin of life. During the Hadean Eon the Sun was substantially dimmer with around ~70% of today's luminosity and with a higher fraction of high-energy UV than during the present Eon. The UV level at the surface would have been much higher due to the absence of ozone shielding, with a minor effect from stellar evolution. The dimness of the Sun impacted the composition of the early atmosphere, for example leading to a high level of carbon dioxide and potentially other (reducing) greenhouse gases. The high flux of UV radiation probably drove photolytic processes that could have generated molecules crucial to early chemical evolution or proto-metabolism. Other host star types with different spectra and evolutionary histories must be considered when extrapolating "origin of life" scenarios to exoplanetary environments.

1b. Accretionary History and Planetary Dynamics (Rebecca Fischer, Harvard University): It is probable that the Earth received most of its inventory of volatiles late in its growth history, as judged by comparison to the distribution of volatiles in the Solar System. More than 90% of Earth's carbon and sulfur may reside in its metallic core, with greater uncertainty in the core's hydrogen inventory. The redox state of Earth's magma oceans was influenced by the Earth's core, but this is not well understood. These phenomena are important for prebiotic chemistry because, by direct interactions with the atmosphere, the magma ocean had a strong influence on the composition of Earth's earliest atmosphere.

1c. Origin of the Moon (Miki Nakajima, University of Rochester): It is generally accepted that the moon was formed by a giant impactor at the end of the planetary accretion stage. The size and velocity of that impactor are actively debated. Models of the impact help

explain depletion of volatiles of the Moon, small isotopic differences between the Earth and Moon, and the angular momentum of the Earth-Moon system. The Moon-forming event had a strong influence on Earth's early habitability because it ultimately determined the length of the day and influenced the compositions of the Earth's atmosphere, mantle, and core.

1d. Hadean Geodynamics (Richard Carlson, Carnegie Institution for Science): The Earth's bulk composition was established, and core-mantle separation initiated before the Moon-forming impact melted large portions of the mantle. The resulting magma ocean formed Earth's primary crust and crystallized to a compositionally heterogeneous mantle. The formation of a felsic crust (rich in aluminium, silicon, potassium, and sodium) began by 4.37 Gyr. This crust was recycled, and the mantle stirred, erasing much of the evidence for earlier differentiation events. Crustal formation and recycling in the Hadean may have been driven primarily by tectonics, but also by impacts and other sources. Between 4.3 and 3.8 Gyr, Earth's surface was sufficiently cool to support liquid water.

1e. Impact History (Simone Marchi, Southwest Research Institute): The Earth was battered during its first billion years by large impacts, including those leading to the formation of the Earth and Moon and additional collisions by leftover planetesimals. The bombardment between 4.5 and 3.5 Gyr was intense, as judged by highly siderophile elements on the Earth and lunar craters. These collisions affected prebiotic chemistry in many ways, including the delivery of CNS elements, impact melting and mixing of the crust, and the release of large amounts of carbon dioxide via outgassing of impact-melt pools. Further, impact-generated hydrothermal systems were a significant component of Earth's near-surface composition and topography.

Theme 2: Evolution of the Near Surface (Oct 16, 2020).

Theme 2 focused on connections between the near surface environment, which hosts the suite of environments and geologic processes within which life arose, and internal processes within the early Earth. Exploring this connection requires understanding the composition of the primordial crust, mechanisms of crustal generation, chemical and physical weathering, and recycling. Other important inputs include the compositions of the atmosphere, ocean, and hydrothermal fluids interacting with rocks. Theme 2 also emphasized the interplay of early planetary processes. Recordings of these talks can be found here (<https://www.youtube.com/playlist?list=PLvogKQh-bBnXUK-aP4ixooMSyzejRD3G>).

2a. Earliest Crust and Chemical Crustal Evolution (Ann Bauer, University of Wisconsin-Madison): Changes to the composition and volume of continental crust are important influences on the chemical cycling of nutrients and volatiles throughout Earth history. On modern Earth, these processes, in tandem with the production and destruction of continental crust, are largely determined by plate tectonics. Therefore, the timing of the initiation of plate tectonics is critical for understanding how the composition and volume of continental crust has varied with time. The early continental crust, while sparse, is a critical archive for evaluating the geodynamic processes that were operating on the early Earth. This record remains a primary source of direct evidence to evaluate the presence/absence of plate tectonics in the geologic record, changes in crust volume with time, and changes in chemistry of the continental crust across billions of years.

2b. Physical Crustal Evolution (Bradley Foley, Pennsylvania State University): The scarcity of early continental crust available for direct study coupled with a poor understanding of the interior modes that influence surface expressions has led to the need for modeling to better explore the connections between Earth's interior and near surface. The dynamics of

the interior and mode of surface tectonics shaped the evolution of the climate and surface environment. However, there is no consensus as to whether plate tectonics operated on the very early Earth as it does on the modern Earth. Alternatives include a stagnant-lid (~ single plate) regime or ephemeral episodes of subduction. These different tectonic regimes would still allow for the carbonate-silicate cycle to operate and to regulate Earth's early climate. The area of exposed land was likely lower in the Hadean and Archean, due to a hotter and potentially H₂O poor mantle, leading to more voluminous early Earth oceans. However, there may have been some subaerial land, derived from mantle plumes, incipient subduction, and impact-generated topography.

2c. Atmosphere and Ocean Evolution (Kevin Zahnle, NASA Ames Research Center): The carbonate-silicate cycle and the ability to regulate Earth's climate is deeply connected with the composition of the oceans and atmosphere. Possible Hadean climates range from too hot for life to surfaces covered in ice, and every climate in between. Atmospheric chemistry presents an apparent paradox: the abiotic chemistry that evolves into life is facilitated by a highly reduced atmosphere rich in methane and highly reduced forms of nitrogen (nitriles or ammonia), but most geological arguments imply that the atmosphere was CO₂-dominated, and its nitrogen available only as N₂ or oxides. The paradox can be resolved by the late accretion of iron-rich material that preferentially and transiently reduces the atmosphere, ocean, and crust.

2d. Lithospheric Fluid Composition (Everett Shock, Arizona State University): The lithosphere is the mechanically rigid outer layer of the Earth, comprising both the upper mantle and the continental and oceanic crusts. Deep fluids, over wide ranges of temperature and pressure, circulate through fractured and permeable rocks, driving weathering and alteration, and facilitating the transfer of electrons from the lithosphere to the near-surface hydrosphere. Interactions between deep fluids and lithospheric crust can produce H₂ that might have driven abiotic synthesis of organic compounds for insertion into prebiotic pathways. Alteration of both intrusive and extrusive ultramafic rocks (serpentinization) can produce more H₂ than mafic picrites and basalts, however even in basaltic mid-ocean ridge settings significant H₂ production is expected in systems with higher temperatures and lower water:rock ratios. Production of H₂ and subsequent abiotic synthesis by deeply circulating fluids could have supported emergence of life, driven by internal lithospheric processes.

Theme 3: Inventories, Geological Settings, and Building Blocks (Oct. 23, 2020).

Theme 3 explores the production of organic compounds essential to the origin of life and how the fate of these compounds was related to specific atmospheric conditions, geologic settings, and local environments. Possible origins of prebiotic organic material include exogenous delivery from meteorites and other impactors, atmospheric chemistry, and fluid-rock reactions at and near the surface. Recordings of these talks can be found here (https://www.youtube.com/playlist?list=PLvogKQh-bBnWx-eZBhRDf1ILLT_XeACFb).

3a. Geological Settings and Local Conditions (Martin Van Kranendonk, UNSW Sydney): Uncertainty about conditions on the early Earth arises in part from a sparse rock record prior to 4.03 billion years ago (Ga). However, extrapolation from younger terrestrial rocks and planetary neighbors has been used to provide guidance. The ancient crust of Mars, for example, shows predominantly basaltic surface rocks (rocks poor in silica but enriched in Mg and Fe when compared to Earth's modern day continental crust) that are pockmarked by meteorite impacts. Some have suggested that early Earth crust would have had compositional similarities to this ancient Martian crust, which is still largely preserved for study. However,

there is not yet community consensus for this model. The raining out of the oceans 4.2 billion years ago would have led to widespread water-rock interactions generating interesting chemistry at spreading centers and in impact basins. Ongoing impacts and rock recycling would have begun differentiation of the crust to granitic compositions, leading to further geochemical complexity. By at least 3.7 Ga, there would have been exposed land surfaces, with preserved hot spring settings by at least 3.5 Ga also providing new geological settings for prebiotic chemistry.

3b. Exogenous Delivery of Organic Matter in the Solar System (Zita Martins, Instituto Superior Técnico, Portugal): The organic content of primitive solar system bodies, as understood through laboratory analyses and space missions, provides important information about the chemistry of the early Solar System and the formation of the Earth. Comets, asteroids, and carbonaceous meteorites delivered exogenous organic matter to the early Earth, and these organic materials are resources on the early Earth and other solar system bodies necessary for the origin of life. Many key compounds in terrestrial biochemistry (e.g., amino acids, nucleobases, and sugars) have been identified as extraterrestrial compounds in carbonaceous meteorites. Investigations have also shown that hypervelocity impact shock of a typical cometary ice mixture produces amino acids, providing a further source of prebiotic organic matter.

3c. Haze and Atmospheric Synthesis (David Catling & Nicholas Wogan, University of Washington): Atmospheric chemistry is a plausible source of organic molecules and required precursors (“feedstocks”) for chemical evolution. In particular, atmospheric chemistry can produce nitriles such as HCN and cyanoacetylene, which potentially react to make complex molecules such as nucleobases. This photochemistry requires a reducing atmosphere similar to modern Titan, which is rich in N_2 with some CH_4 . Although the dominant Hadean atmosphere was likely CO_2 - and N_2 -rich, intermittent conversion to highly reducing atmospheres would have been triggered by large impactors. Iron from impactor cores would have reacted with seawater to create H_2 , which can reduce CO_2 to CH_4 . Because hydrogen gradually escapes to space, such reducing atmospheres would persist for tens of thousands to millions of years, scaling with the size of the impactor. Impacts large enough to make thick H_2 atmospheres (i.e., at least ~ 1 bar H_2) probably occurred on a repeat timescale on the order of ~ 10 s Myr during the Hadean, with smaller cadences in the early Hadean. Feedstock molecules made by atmospheric chemistry, such as cyanides, could be sequestered and concentrated for prebiotic schemes, and closed-basin lakes or pools provide such a mechanism. Recent experiments and models have shown that sodium carbonate lakes favor concentration of cyanides at pH values consistent with laboratory-demonstrated prebiotic reactions. The same lake chemistry also concentrates phosphate sourced from weathering. Such lakes provide a means for cyanide and phosphate to accumulate to levels needed for organic synthesis.

3d. Surface Chemistry and Abiotic Organic Synthesis (Bénédicte Ménéz, Université de Paris): The Earth’s lithosphere can abiotically generate organic compounds through geological processes and associated fluid-rock reactions that produce chemical disequilibria. Redox reactions play a pivotal role in abiotic formation of organic compounds and transformations associated with magmatic, tectonic, hydrothermal, and radiolytic processes, although reaction pathways and product diversity are still unconstrained. These reactions are also influenced by minerals and dissolved metal ions that can enhance reaction rates and selectively control reaction pathways. An important role is played by microenvironments characterized

by steep redox and chemical gradients that are expected to evolve with time, tectonics, and hydrodynamics. Geologic hydrogen (H_2), widely produced on Earth throughout its history, plays a central role in these processes that today sustain subsurface chemosynthetic biological communities, and is likely key to understanding the conditions that ultimately led to the emergence of life on Earth and possibly elsewhere in the solar system.

Theme 4: Prebiotic Complexity (Nov. 13, 2020).

Theme 4 was focused on the formation of small molecules and polymers important for the origin of life. In addition to their prebiotic synthesis, a central thesis of this theme was molecular interactions and possible function before the onset of life. Recordings of these talks can be found here (<https://www.youtube.com/playlist?list=PLvogKQh-bBnUgZJ0scf1nLkMKFjPDS1I8>).

4a. Overview (David Deamer, University of California, Santa Cruz): The sources of organic compounds and forms of available energy are linked to the role of water and self-assembly processes. Evolution of protocells is driven by the synthesis of polymer systems and their encapsulation. Encapsulation of polymers can act as important step for selection and evolution.

4b. Precursors, Simple Molecules, and Selection, Part 1 (H. James Cleaves, Earth-Life Science Institute): A variety of organic and inorganic chemicals may have been available on the primitive Earth. Their identities and abundances are a matter of open discussion and would have depended on specific atmospheric conditions and surface geochemical conditions. It is possible that evolvable prebiotic systems were very different from modern biochemical systems. It is important to understand production and properties of molecules in geochemical reaction context. Their emergence was limited by parameters defined by physical chemistry, geochemistry, and the nature of complex unguided chemical systems.

4c. Precursors, Simple Molecules, and Selection, Part 2 (Laurie Barge, Jet Propulsion Laboratory): Prebiotic organic reactions on early Earth may have formed in the presence of minerals and inorganic ions, guided by the geological environment. Minerals and organic compounds impact one another in various ways that are relevant for origin of life: for example, minerals can catalyze organic synthesis, and organic molecules can trap minerals in metastable, reactive phases. All these factors together can drive various types of organic selection, e.g., partitioning organic compounds in liquid phases and adsorbing them on mineral surfaces, adsorption/desorption of organics can be driven by changes in pH or competing ions, and/or organic redox chemistry of reactive metals and minerals.

4d. Processes Acting on Building Blocks, Assembly and Complexification, Part 1 (Luke Leman, Scripps Research): Rapid advances are being made in experimental production of proto-polymers. It is thought that prebiotic polymers of some sort played an enabling role in the transition from nonliving matter to life. The ultimate conclusion of this transition was the initial formation of proteins, nucleic acids, and carbohydrates. Abiotic polymerization reactions can be promoted by thermal processes, hydration-dehydration cycles, and chemical activation of the building blocks, especially if coupled with a mechanism for organizing the building blocks along surfaces, interfaces, or templating molecules. A major unresolved challenge is to better explore the consequences of heterogeneous building block mixtures on abiotic polymerization reactions and the resulting heterogeneous product polymers. Although oligomers produced in known abiotic processes are typically much shorter than polymers found in today's biochemistry, the assembly of oligomers into noncovalent com-

plexes could have aided in bootstrapping up more complex structures and functions from relatively simple oligomers.

4e. Processes Acting on Building Blocks, Assembly, and Complexification, Part 2 (Christine Keating, Pennsylvania State University): It is thought that prebiotic complexity increased upon production of polymers and amphiphiles capable of self-assembly. Polymers fold to adopt three-dimensional conformations and interact with other molecules of the same or different types. Intra- and intermolecular self-assembly provides increased chemical stability and new functions such as catalysis, recognition, and possibly self-replication. Several types of molecular self-organization can generate compartments, including amphiphile self-assembly to form micelles or vesicles and a type of polymer phase separation called coacervation. Prebiotic compartments could have provided favorable microenvironments and been critical steps in protocell formation. Selective pressure can act on populations of assembled molecules or compartments, leading to changes in composition and chemical evolution. Synergies arising from co-assemblies of different molecular classes and assembly types can be realized. Favorable conditions for increasing complexity via self-assembly could include environmental factors such as gradients and cycling.

Theme 5: Peering into the Past with Today's Biochemistry (Nov. 20, 2020).

Theme 5 focused on the transition from prebiotic chemistry to the first living entities from several different perspectives, including discussions of the nature of chemical evolution, descriptions of chemical model systems, and the search for fossils of early life forms. Among the key points emphasized was a reminder that the tendency to assign a central molecule to the emergence of life—nucleic acids, metabolites, and peptides—may be a misleading extrapolation back from today's biology. Recordings of these talks can be found here (<https://www.youtube.com/playlist?list=PLvogKQh-bBnWtoC4qvIR1PHly0OL6v3KU>).

5a. Overview (Ram Krishnamurthy, Scripps Research): Efforts to unravel the transition of prebiotic chemistry into biochemistry, leading to life on Earth, frequently take clues from, and rely substantially on, extant biochemistry. This focus on biomolecules and biological phenomena deemphasizes the diversity of the prebiotic molecular inventory and the potential creativity of chemical evolution. Our understanding of prebiotic syntheses of different classes of biomolecules suggests that extant biochemistry has in many cases lost some connections to prebiotic synthesis. As a result, stand-alone models (such as the RNA or metabolism worlds) appear inadequate. We suggest a more holistic and interdisciplinary approach to the origins of life that considers co-evolution of early Earth environments with prebiotic processes and pathways.

5b. Genetics (Hannes Mutschler, Max Planck Institute of Biochemistry, Martinsried): All known living organisms require a genetic system to ensure survival and evolutionary adaptation. The basic properties and replication mechanisms of nucleic acid-based genetic systems direct the flow of information in all life forms. However, alternative "artificial" systems have been discovered for encoding genetic information. In this context, there are different variations of the RNA-world hypothesis. The relationship between environmental conditions and the stability of nucleic acid components may have influenced the emergence of the first genetic systems.

5c. Metabolism (George Cody, Carnegie Institution for Science): A flux of simple organic and inorganic substrates pass through core metabolic pathways of extant biology. It seems likely that dynamic, environmentally driven organic reaction networks preceded the origins of life on Earth. Sustainable dynamic organic reaction networks can lead to molecular con-

stituents familiar to us in the core of intermediary metabolism. The apparent ubiquity of the molecular constituents of such networks is important both in the context of natural systems (planetesimal interiors of carbonaceous chondritic parent bodies) and in experiments exploring transition metal sulfide-catalyzed aqueous organic reactions.

5d. Chemical Evolution (Moran Frenkel-Pinter, Georgia Institute of Technology): While it may not be possible to recapitulate the emergence life, it is possible to understand the chemical principles that led to it. We consider chemical evolution to be a process of continuous molecular change, with progression to new chemical spaces. In our model, early stages of chemical evolution selected monomers based on kinetic and thermodynamic landscapes governing condensation and hydrolysis. Intermediate stages selected oligomers on the basis of emergent properties such as folding, assembly, and catalysis. The end phases gave rise to biopolymers. Chemical evolution harvests energy provided by environmental cycling, including wet-dry cycling. A near-equilibrium chemical mixture that uses water as both reactant and product can chemically ratchet, when subjected to oscillating water activity.

5e. Search for the Earliest Evidence of Terrestrial life (Elizabeth Bell, University of California, Los Angeles). The search for the earliest evidence of life must include three perspectives: whether a structure is biological (textural, chemical, and isotopic), how well the age of the host rock can be established, and whether the structure is primary to its host rock or can otherwise be directly dated. The search for the earliest evidence of terrestrial life is mainly limited by the sparseness and alteration of the ancient rock record. Around 0.5 billion years separate planet formation and a semi-continuous rock record. Increasing degrees of metamorphism in the rock record with increasing age obscure or destroy evidence of a primitive biosphere. Nevertheless, investigations have uncovered structures resembling microfossils and stromatolites (microbial mats) in the metasedimentary record dating to nearly 3.5 Ga, along with light carbon isotopic measurements of graphite included in older crystals of apatite and zircon.

5 f. Phylogenomic Extrapolation from Existing Life to Early Life Forms (Greg Fournier, Massachusetts Institute of Technology). Genome sequence information conserved across the Tree of Life potentially contains information about the earliest divergences of life's history. This information can be used to generate lineage phylogenies that reach back to LUCA, the Last Universal Common Ancestor, showing that "modern" core molecular biology was already in place by the time of LUCA. A small number of protein families are shown to have diversified pre-LUCA and are an invaluable and unique source of information about the earliest life. Molecular clocks can provide younger bounds on the timing of the deepest divergences of life, and ancestral sequence reconstruction can provide insight into the composition of the earliest proteins and provide clues to their environment. All of these inferences are limited by inherent phylogenetic uncertainty, information loss, and the complex evolutionary history of gene families.

Summary and Future Prospects

The workshop was structured through talks and breakout sessions to explain wide-ranging ideas and efforts of the community and to identify questions, answers, opportunities, and impediments. Insights and questions from the breakout discussions are summarized below.

Prebiotic chemistry was influenced by the Earth's accretion and early evolution (Wilde et al. 2001; Kramers 2007; Armstrong et al. 2019). Throughout the Hadean, the atmosphere, ocean, and lithosphere co-evolved with the internal state of the planet. The system was shaped by large impactors (Canup 2004; Nakajima and Stevenson 2015) and the infall of smaller meteorites (Abramov et al. 2013; Kadoya et al. 2020). Impactors may have periodically generated reducing atmospheric conditions with time constants of millions of years (Benner et al. 2020; Zahnle et al. 2020). The orbital configuration of the Moon led to higher frequency of day-night cycles than today (Canup 2004) and more intense tides. Questions remain about the volume and composition of early oceans and the abundance of dry land (Korenaga 2021), whether generated by plate tectonics, impacts, or plume volcanism.

The conditions, chemical components, and processes that favored the rise to life are intensely debated and remain unresolved. The chemical environments at and below the surface were characterized by wide-ranges in pH, water activity, ion concentrations, temperature, reductive potential, and pressure (Sojo et al. 2016; Halevy and Bachan 2017; Westall et al. 2018; Barge et al. 2019; Ross and Deamer 2019; Damer and Deamer 2020; Zahnle et al. 2020). Possible factors contributing to formation and assembly of early life's building blocks, oligomers and proto-polymers are wet-dry cycles (Forsythe et al. 2015; Frenkel-Pinter et al. 2019; Damer and Deamer 2020), freeze-thaw cycles (Mutschler et al. 2015; Lie et al. 2016), temperature-pressure variation, fluid flow, water-rock reactions and chemical gradients (Baross and Hoffman 1985; Martin and Russell 2007; Martin et al. 2008), atmospheric turbulence, and volcanic outgassing (Griffin et al. 2018). Despite the many details under debate, it is broadly accepted that the dynamic nature of local and global environments, including recurrent and periodic variations with diverse time constants, could have played important roles in prebiotic chemistry.

Workshop participants were interested in factors critical to the prebiotic synthesis of organic compounds. Organic molecules were produced on the ancient earth by weighted contributions from (i) delivery by meteorites and comets (Schmitt-Kopplin et al. 2010; Abramov et al. 2013; Mehta et al. 2018; Kadoya et al. 2020), (ii) synthesis in the atmosphere (Miyakawa et al. 2002) in processes driven by solar irradiation (Lingam et al. 2018), and (iii) generation on the surface and the subsurface (Sojo et al. 2016; Westall et al. 2018; Ross and Deamer 2019; Damer and Deamer 2020), via (iv) reactions catalyzed by minerals (Wilde et al. 2001; Takeuchi et al. 2020). The earliest forms of catalysis may have relied on mineral surfaces and/or metal ions (Ferris 2006; Cleaves II, et al. 2012) or organic molecules (Forsythe et al. 2015; Frenkel-Pinter et al. 2019) to mediate chemical processes such as oxidation-reduction or condensation-dehydration. Workshop participants noted the importance and possible interdependence of parameters and the lack of constraints in current models. Understanding the origins of life requires knowledge of prebiotic chemical pathways, including input molecules, ranges of products and steady-state abundances.

Some breakout exchanges focused on possibilities of proto-metabolic pathways, which may have been catalyzed (Morowitz et al. 2000; Hordijk et al. 2010; Goldford et al. 2017; Yadav et al. 2022). Compartments and molecular sequestration were discussed. In addition to serving as evolutionary units, compartments may have helped to concentrate molecules and mediate specific reactions. The earliest compartments could have been some combination of vesicles (Lopez and Fiore 2019), coacervates (Cakmak et al. 2020), mineral pores (do Nascimento Vieira et al. 2020), and viscous surface systems (He et al. 2017). The types and

functions of compartments and their possible variation during the progression from abiotic to biotic world were identified in breakout sessions as important topics for investigation.

Life requires energy. Discussions focused on the earliest forms of proto-biological energy (Scharf et al. 2015), which might have been in the form of thermal (Forsythe et al. 2015; Frenkel-Pinter et al. 2019; Ross and Deamer 2019; Damer and Deamer 2020) or chemical (Wächtershäuser 1988; Guillot and Hattori 2013; Barge et al. 2017) or mechanical energy (Bolm et al. 2018; Steele et al. 2020), with possibilities of activated compounds generated via solar energy (Lingam et al. 2018), or delivered by meteorites (Chyba and Sagan 1992). Integration of the processes described above may require a “systems chemistry” approach (Ruiz-Mirazo et al. 2014; Lopez and Fiore 2019).

The earliest forms of assembly, catalysis, and/or replication may have required homochirality and chemical homogeneity (Ribó et al. 2017; Burton and Berger 2018; Lee et al. 2022). The breakout sessions discussed uncertainties about how enantiomeric excesses observed in biology arose. Important questions include when, and how chiral and chemically homogeneous building blocks were selected.

The nature of the transitions from chemistry to chemical evolution to Darwinian evolution were discussed in the breakout sessions. An important and controversial topic is the nature of chemical progression and primitive evolution prior to establishment of the Central Dogma of Molecular Biology, which involves DNA, RNA and polypeptide. In some models, a single proto-biopolymer capable of catalysis, information storage and self-replication is an important stepping stone to current biology (Bernhardt 2012; Benner et al. 2020). In a subset of these models, purely abiotic processes, in the absence of chemical evolution, produced nucleotides, which polymerized to provide the first RNA polymers (Dworkin et al. 2003; Neveu et al. 2013), initiating Darwinian processes. In other models, RNA is the product of evolution of a more primitive proto-nucleic acid (Hud et al. 2013). In most RNA-centric models, the ribosome and protein synthesis are late addenda to proto-biology. An alternative class of models proposes instead that a creative stage of chemical evolution (Yu et al. 2016; Frenkel-Pinter et al. 2019, 2020; Stolar et al. 2021) merged smoothly with Darwinian processes. In some of these models the ribosome was an early foundation to proto-biology; multiple types of biopolymers, some primarily informational and others primarily catalytic, arose in concert via co-evolution (Lanier et al. 2017; Frenkel-Pinter et al. 2020).

During discussions it was noted that processes that were central in the emergence of life may not have persisted into modern life. It is possible that intense creativity of early evolutionary processes (Jacob 1977) sufficiently erased chemical history (Gould and Vrba 1982; Gould 2002) to the extent that modern biology is not informative about specific chemical species and processes during the earliest stages in origins of life.

Early biological systems did adapt and persist. Workshop participants speculated on the nature of early systems and on how such systems could preserve essential functions during environmental change. The First Universal Common Ancestor (FUCA) and the Last Universal Common Ancestor (LUCA) are hypothetical organisms or consortia of organisms (Penny and Poole 1999; Koonin 2003; Weiss et al. 2016) that remain to be fully understood. The biochemical makeup of LUCA can be partially inferred from the features that are now distributed over all domains of life (Koonin 2003; Bernier et al. 2018). Extrapolating to FUCA and to earlier stages (Fournier et al. 2011; Petrov et al. 2014, 2015) – with all the care that must be taken with this bio-centric approach – may allow us to tease out clues about the molecular organization of the earliest proto-biological life forms and their interactions

with their environments. Discussions also addressed “minimal living systems” (Forster and Church 2006; Caschera and Noireaux 2014) and speculated on their relationships to the origins of life.

Many of the topics discussed in the symposium and breakout sessions are controversial, and some researchers are highly invested in specific models. As expected, there were points of disagreement, but these often emerged as topics of constructive debate. Steady progress is being made in understanding our chemical and biochemical place in the universe. The question of the origin of life is an important and solvable problem that requires additional information on early Earth environments.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s11084-022-09622-x>.

Declarations

Conflict of Interest The authors state that they have no conflicts of interest either directly or indirectly related to this work.

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Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

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