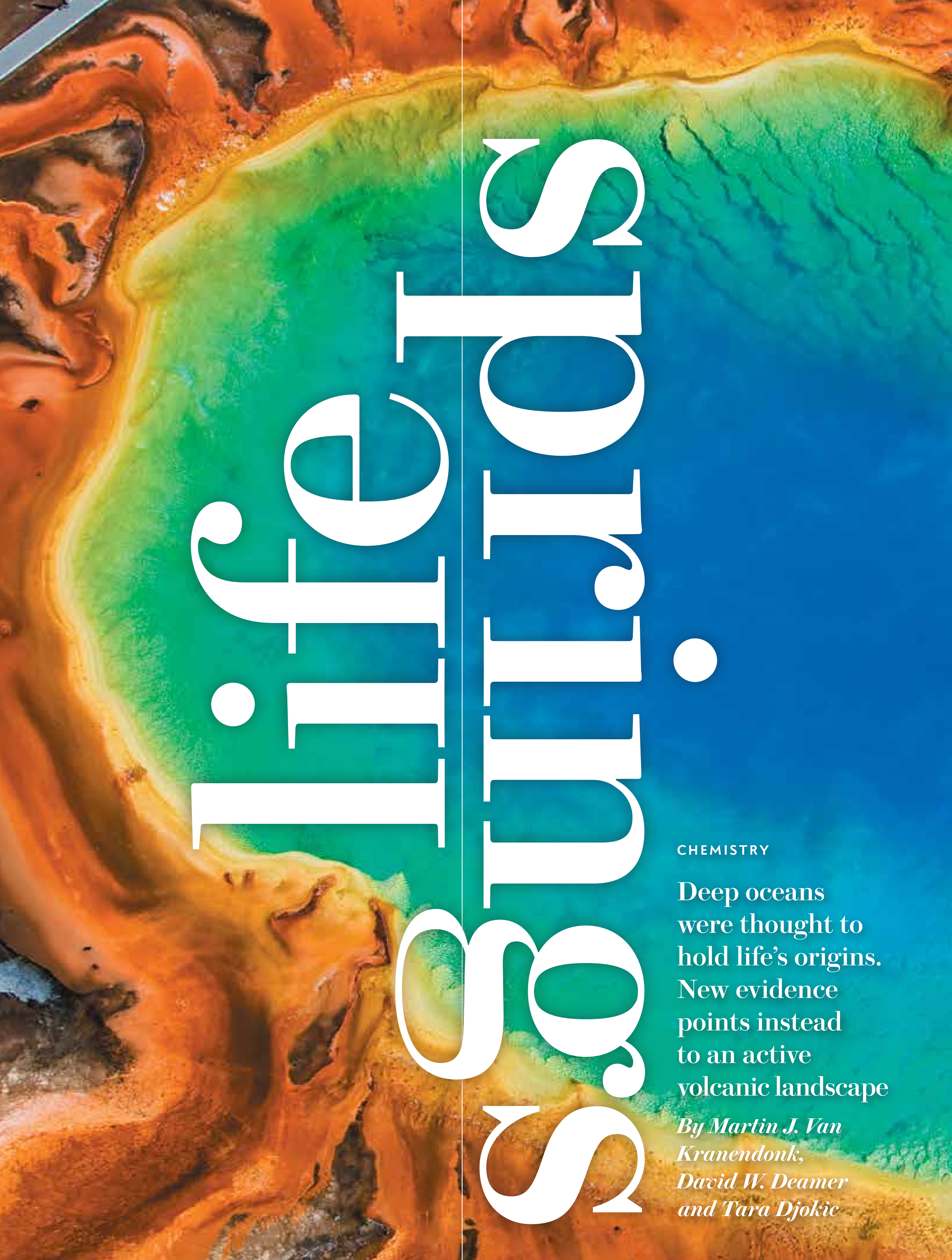

CHEMISTRY

life springs

Deep oceans were thought to hold life's origins. New evidence points instead to an active volcanic landscape

By Martin J. Van Kranendonk, David W. Deamer and Tara Djokic

BIRTHING POOL: Life on Earth could have started in places similar to the Grand Prismatic Spring in Yellowstone National Park.

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David Deamer is a faculty member in the department of biomolecular engineering at the University of California, Santa Cruz. He is author or editor of 12 books, including *The Origins of Life* (2010), co-edited with Jack W. Szostak, and *First Life* (2011), published by the University of California Press.

Tara Djokic is a Ph.D. candidate at the Australian Center for Astrobiology at the University of New South Wales. Her project combines geologic observations of early evidence of life in Western Australia with virtual-reality technology.

IT'S PITCH-BLACK. WE HAVE BEEN SCRATCHING OUR WAY THROUGH DENSE UNDERBRUSH IN northwestern Australia, guided only by the dim light from a GPS screen. The light is too weak to reveal fallen trees that fill the dry creek bed we are following, and we keep tripping over them. We are two geologists working in a remote region of the country known as the Pilbara: Djokic up front and Van Kranendonk several steps behind. Our truck, parked somewhere on a small plateau, seems a world away. We are not sure if the GPS's batteries will hold out long enough to show us the way back. The night sky, ablaze with countless stars visible right down to the horizon, twinkles in an amazing spectacle as Jupiter dances with nearby Venus. Sadly, this spectacle provides little navigational help for two scientists fumbling their way through the Australian outback in June 2014.

Heading up the side of the creek embankment, Djokic suddenly stumbles back downhill. Has she lost her balance? To stop her from falling, Van Kranendonk reaches out to stop her and pushes her back uphill, which prompts a screech, something unintelligible, and finally a sputtered cry: "Sp- … p- … p- … pppider!" Djokic has not stumbled at all. She is in flight mode, in fear for her life as she tries to swat away the thick spider web enveloping her. Spiders have a deservedly bad reputation in Australia. In the dark, it is not a good idea to assume that you have found the odd benign species.

The reason we are feeling our way around the Pilbara at night is because we had spent the day enthralled by a new discovery Djokic had made in 3.48-billion-year-old sedimentary rocks called the Dresser Formation. Some of the rocks are wrinkled orange and white layers, called geyserite, which were created by a volcanic geyser on Earth's surface. They revealed bubbles formed when gas was trapped in a sticky film, most likely produced by a thin layer of bacterialike microorganisms. The surface rocks and indications of biofilms support a new idea about one of the oldest mysteries on the planet: how and where life got started. The evidence pointed to volcanic hot springs and pools, on land, about 3.5 billion years ago.

This is a far different picture of life's origins from the one scientists have been sketching since 1977. That was the year the research submarine *Alvin* discovered hydrothermal vents at the bottom of the Pacific Ocean pumping out minerals containing iron and sulfur and gases such as methane and hydrogen sulfide, surrounded by primitive bacteria and large worms. It was a thriving ecosystem. Biologists have since theorized that such vents, protected from the cataclysms wracking Earth's surface about four billion years ago, could have provided the energy, nutrients and a safe haven for life to begin. But the theory has problems. The big one is that the ocean has a lot of water, and in it the needed molecules might spread out too quickly to interact and form cell membranes and primitive metabolisms.

Now we and others believe land pools that repeatedly dry out and then get wet again could be much better places. The pools have heat to catalyze reactions, dry

PRECEDING PAGES: WERNER VAN STEEN *Getty Images*

IN BRIEF

To get started, life on Earth needed energy to create complex molecules and ways to bring these molecules together.

A system of volcanic pools and hot springs on land has the needed ingredients for life and wet-dry cycles for interaction and natural selection.

A land-based volcanic origins theory, in contrast to an ocean-focused one, guides us to different places in the solar system to search for life there.

spells in which complex molecules called polymers can be formed from simpler units, wet spells that float these polymers around, and further drying periods that maroon them in tiny cavities where they can interact and even become concentrated in compartments of fatty acids—the prototypes of cell membranes.

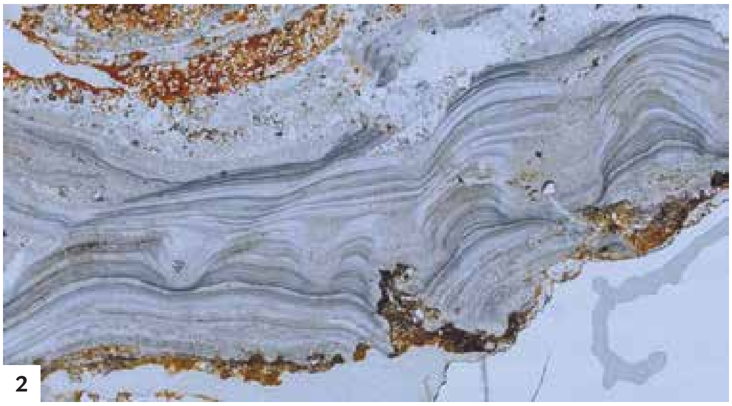
What Djokic found was strong geologic evidence that the Dresser, now a dry, hot and barren outback environment, had once been like the steaming pools and erupting geysers of Yellowstone National Park in the U.S., an active geothermal field. And everywhere in the Dresser there are fossilized signs of life intimately associated with the old hot spring system. Although the Dresser was not the actual site where the most primitive life began half a billion years earlier, it was showing us that hydrothermal environments on land were present very early in Earth's history. Charles Darwin had suggested, back in 1871, that microbial life originated in "some warm little pond." A number of scientists from different fields now think that the author of *On the Origin of Species* had intuitively hit on something important. And the implications of these ideas stretch beyond our own planet: in our search for alien life elsewhere in the solar system, a land-based theory about origins would guide us to different places and planets than would an ocean-based theory.

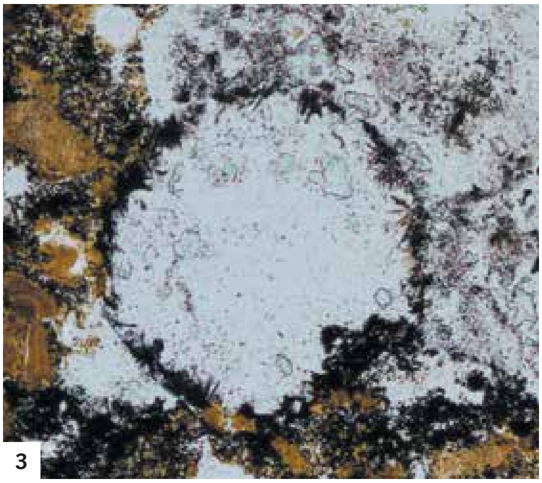
FROM RUSSIA WITH LIFE

TEN YEARS BEFORE Djokic's run-in with the spider web, another of us (Deamer) had shown that volcanic pools could foster the assembly of compartments made of membranes, essential boundaries of all cellular life. Deamer led a group of scientists to Mutnovsky, an active volcano in the Kamchatka peninsula of far eastern Russia. The group was exploring a prebiotic analogue site, a region that can give researchers a sense of what the planet was like four billion years ago, before life began. Deamer's idea was that simple molecular building blocks might join into longer information-carrying polymers like nucleic acids—needed for primitive life to grow and replicate—when exposed to the wet-dry cycles characteristic of land-based hot springs. Other key polymers, peptides, might form from amino acids under the same conditions. Crucially, still other building blocks called lipids might assemble into microscopic compartments to house and protect the information-carrying polymers. Life would need all the compounds to get started, and Mutnovsky had an abundance of hot springs and geysers in which the idea could be tested.

Deamer had brought a bottle of white powder containing raw material that was likely available on the prebiotic Earth, including four amino acids and four chemical bases that compose naturally occurring nucleic acids, as well as phosphate, glycerol and a lipid. He poured this mixture into the center of a small, boiling spring. Within minutes a white, frothy foam emerged around the spring's edges. The foam was composed of countless tiny vesicles, each containing compounds that were present in the original soup.

COURTESY OF MARTIN J. VAN KRANENDONK





LIFE ON THE ROCKS: Orange rocks in Australia's Pilbara region are called geyserite, composed of minerals splashing from geysers in hot springs (*1*). The rocks show signature dark bands rich in titanium and light bands composed largely of potassium in a microscopic view (one centimeter in width) (*2*). Minuscule bubbles preserved in this 3.5-billion-year-old geyserite were formed in sticky biofilms, the products of biological organisms (*3*).

If the compartments dried out around the edges of the puddle, could their contents, already in close proximity, join together as polymers? Could this be a stepping-stone to the first life? Back in his laboratory, Deamer and his colleagues tested the idea by mixing simple nucleic acids called nucleotides with lipids. The mixture was put through cycles of wetting and drying under the acidic conditions and high temperatures found in the Kamchatka pool. The result: longer polymers ranging from 10 to more than 100 nucleotides in length. Later studies using x-ray diffraction demonstrated the polymers resembled ribonucleic acid, or RNA. Furthermore, these polymers were encapsulated by the lipids to form vast numbers of microscopic compartments called protocells. Though not alive, they were clearly an important step toward life.

Deamer used just a few wet-dry cycles in his experiments and got relatively simple molecules. A colleague of his at the University of California, Santa Cruz, computer scientist Bruce Damer, suspected that many more cycles might add another key feature: the survival of the fittest. Each drying cycle, Damer figured, would cause lipid membranes of the vesicles to open, allowing polymers and nutrients to mix. On rewetting, the lipid membranes would reencapsulate different mixtures of polymers, each mixture representing a kind of natural experiment. More complex protocells would have better chances of survival because their greater variety of molecular mixtures might stabilize the protocells in various conditions—one set of molecules helping in one set of surroundings, another helping in a different set. These intact protocells would then survive to pass on these polymer sets to the next generation, climbing an evolutionary ladder. Damer realized that this model resembled a kind of chemical computer "booting up" the functions of life, starting with random "programs" written in the form of polymers.

In 2015 Damer added a third phase to the two-part cycle: an intermediate stage between wet and dry. The idea occurred during a field trip with the co-authors to the Dresser Formation in search of stromatolites, which are the fossilized layers of bacterial mats and some of the earliest evidence of life on Earth. Damer was walking through the desert near a granite outcrop known as Gallery Hill that is covered with Aboriginal rock carvings known as petroglyphs. On the way, he noticed brown, dried-up microbial mats in small depressions in the outcrops. Out of curiosity, Damer poured water on the mats, and they sprung back to life, becoming green and gel-like. He realized that if wet-dry cycles in an origin pool also included a moist phase, in which surviving protocells crowd together into a similar gel, polymers and nutrient molecules could mix and exchange across the barriers of lipid membranes. This community of cooperating protocells would have even more opportunities to find the best molecules for survival. Forty years earlier, in fact, scientists George Fox and the late Carl Woese proposed the term "progenote" for such a communal primordial phase of life; Fox told Damer this matched his protocell gel.

POOLS OF INNOVATION

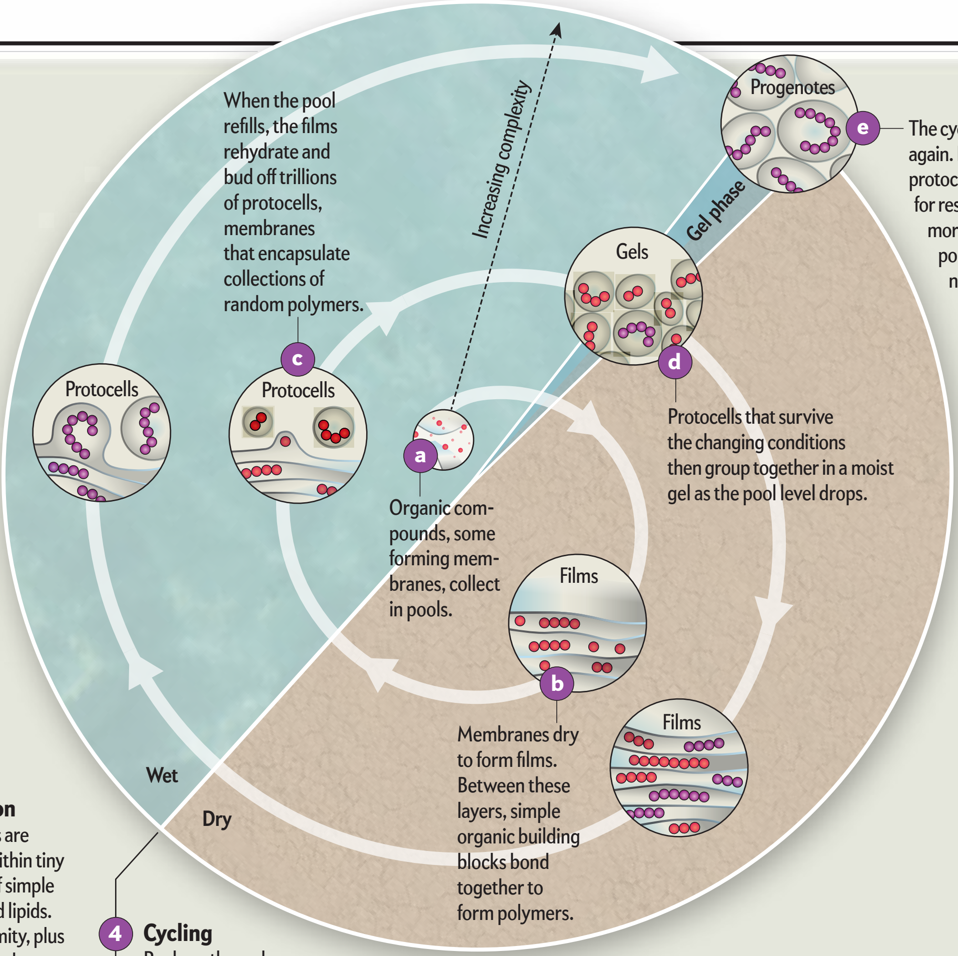
THE BUBBLES AND MINERAL composition that Djokic found in the Dresser Formation made it a likely spot for the three-part cycle to occur, and we published the evidence this past May in *Nature Communications*. After we realized that the Dresser had been filled with surface hot springs in a geothermal system, it became clear that it also had contained many of the key ingredients and organizational structures required for the origin of life. It had a source of energy in the form of

Genesis Landscape

Hot springs, pools and geysers can kick-start chemical systems necessary for life on Earth to begin, according to one theory. The conditions set in motion seven steps, beginning with chemical synthesis, moving through cycles of increasing complexity and ending in colonization of new territory.

1 Synthesis
Many of life's basic building blocks, such as amino acids, are formed in space and fall to Earth.

2 Accumulation
In-falling organic compounds, along with others generated within hot springs on a volcanic landscape, accumulate in hydrothermal pools.



a Organic compounds, some forming membranes, collect in pools.

b Membranes dry to form films. Between these layers, simple organic building blocks bond together to form polymers.

c When the pool refills, the films rehydrate and bud off trillions of protocells, membranes that encapsulate collections of random polymers.

d Protocells that survive the changing conditions then group together in a moist gel as the pool level drops.

e The cycle repeats, again and again. Every time it does, protocells interact, compete for resources and evolve more complex functional polymers until a “progenote” community emerges that is able to exchange adaptive molecules, developing ever more sophisticated functions.

3 Concentration

The compounds are concentrated within tiny vesicles made of simple molecules called lipids. The close proximity, plus heat and chemical energy from the spring system, links them together to form more complex molecular chains.

4 Cycling

Pools go through repeated cycles of three phases: dry, wet and moist gels. Dry times help to synthesize polymers used to carry information, such as chains of nucleic acids. In a wet period, protocells can form, encapsulating these polymers and protecting them. Then, in the gel phase, protocells pack together in a system called a progenote and exchange sets of polymers, selecting those that enhance survival during many cycles.

5 Distribution

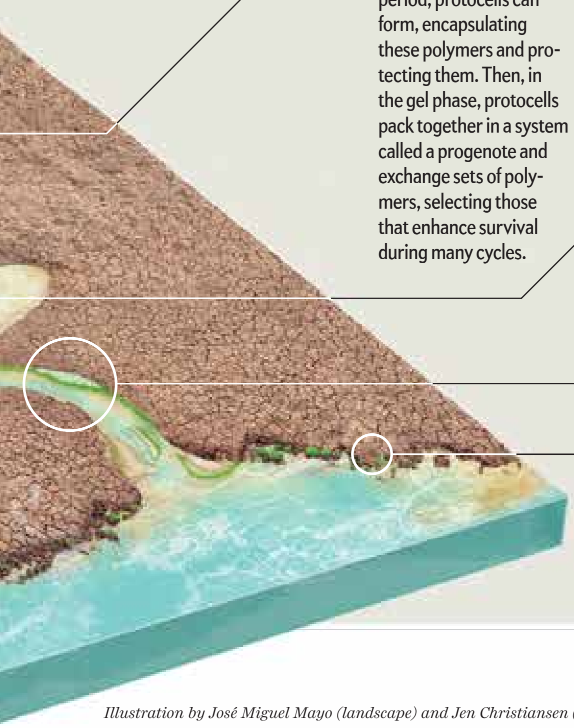
The best-adapted protocells spread to other pools or streams, moving by wind and water, and some develop the ability to use carbon dioxide for photosynthesis. After much trial and error, one protocell assembles the complicated molecular machinery that enables it to divide into daughter cells. This paves the way for the first living microbial community.

6 Adaptation

Some of these early microbes are pushed into saltwater estuaries, beyond their native freshwater ponds. The microbes that survive pass along useful traits that help descendants expand their range to oceans.

7 Colonization

Sea storms and tugging tides select for mats of rugged microbes able to cement themselves together using grains of minerals. These layers pile up into stacks called stromatolites. Life continues to expand into other niches, setting the stage for free-living cells. After billions of years, these organisms evolve into complex multicellular plants and animals.



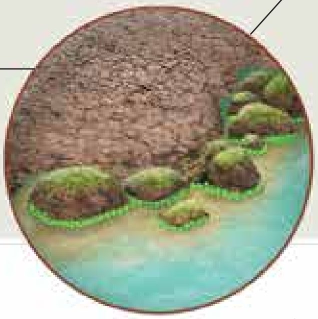


Illustration by José Miguel Mayo (landscape) and Jen Christiansen (cycling detail)

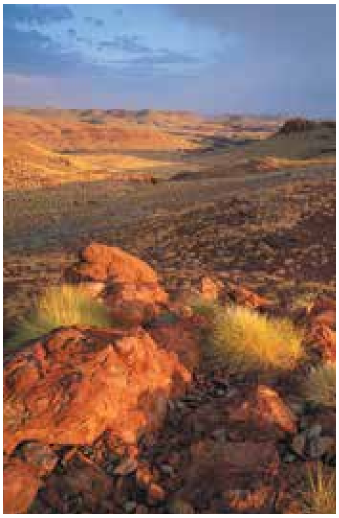
Journey to a Land across Time

When I first set foot in Western Australia's Pilbara, a landscape holding 3.5-billion-year-old clues to the beginning of life, I was very disappointed. The year was 1994. I drove excitedly out of the west coast town of Port Hedland, but all I saw for the first 150 kilometers were a few withered, scraggly trees and smoky dust devils traipsing across the burnt, flat plain. I felt desolated. What had I gotten myself into? And the heat!! I'd never experienced anything this brutal before. Or breathed air so thick with biting flies.

But as we continued to head south on the highway to Marble Bar—the hottest town in Australia—some low, broad hills started to rise from the horizon. We started to cross sandy creeks and rivers, including the mighty Shaw, whose banks were garnished with lush-looking coolabah trees, with their distinctive, bright-white trunks.

As we continued down a dirt track into the hills, the burnt plains gave way to grass-covered hummocks. This grass is called spinifex, an amazing but devilish creation. It grows as bushes up to one meter in diameter, with round, fine blades that taper into needle-sharp tips made almost of pure silica. The tips will penetrate through just about any piece of fabric. My supervisor whipped out thick gaiters to protect his legs. But he had failed to inform me of the hazard. Without any gaiters, I was a walking porcupine within minutes—my skin skewered with multiple silica needle tips that broke off and remained in my flesh for months.

The land, ultimately, proved worth the discomfort. Here I was walking over some of Earth's oldest, best-preserved rocks that contain evidence of life from almost the very beginnings of time on our planet. As I looked at some wrinkly structures that lay above the ripples of ancient sediment, I realized I was looking at remnants of our great-, great-, great-grandparents—the precursors to all complex life on Earth!



CRADLE OF LIFE? Australia's Pilbara region, now dry, once held hot springs and geysers.

This area had changed much from when it was first formed 3.5 billion years ago. Back then it would have been a black volcanic land, with no color from vegetation. Over the hills I might have glimpsed a green, iron-rich sea underneath an orange sky heavy with carbon dioxide and devoid of oxygen. Nearby in the landscape I'd come across fields of hot springs, and here I'd start to see some color. There would be stretches of white and yellow and red around bubbling mud pools and splashing geysers, the colors of sulfur, clay and iron. And in some pools and channels, perhaps there would have been strands of beige, red and purple: colonies of heat- and chemical-loving microbes. There might even have been some green from very early photosynthesizing organisms.

If I were able to ride a time machine forward a billion years, I'd see the Pilbara become buried under kilometers of volcanic lavas and sediments; I'd see the landmass move across the face of the globe and run into other pieces of crust, the collisions forming mountain belts. At about 2.5 billion years ago, I'd see the oceans fill with life, the shallow coastal areas occupied by huge reefs made of primitive microbes called cyanobacteria that stack in piles of mats called stromatolites. The sky would turn blue as the photosynthesizing cyanobacteria sucked in carbon and pumped out oxygen into the atmosphere. Almost another two billion years later the world would turn cold and become covered in a global ice sheet, wiping out almost every living thing. When it melted away, oxygen levels rose again. Life really got going. Animals slowly colonized the land, as did new types of plants. The greening of our planet began in earnest, and a wide variety of organisms appeared—including, unfortunately for me, spinifex.

—M.J.V.K.

circulating hydrothermal fluids, rich in hydrogen, heated by magma from below. The rocks contained abundant amounts of the element boron, a crucial ingredient in the synthesis of ribose necessary for nucleic acids such as RNA. The Dresser also has phosphate minerals that dissolve out of the underlying rocks and join circulating acidic geothermal fluids. Phosphate is an important component of nucleic acids, but it is also used by all life in the form of ATP (adenosine triphosphate, the molecule that supplies energy within cells). In addition, there were high concentrations of zinc and manganese, components of many enzymes in the cytoplasm of cells from all known branches of life, found in hydrothermal vents and in evaporative volcanic lake deposits. Finally, the Dresser also had clays, which can function as catalysts for creating complex organic molecules because of the electrically charged layers of mineral surfaces they contain.

Perhaps the most exciting thing about the Dresser as an origin analogue site is its amazing variety because in this field of science, variety is very much the spice of life. The Dresser is dry and rocky now, but in their youth, geothermal hot spring fields such as this one contain many hundreds of pools, each with a slightly different pH, temperature, dissolved ions and other chemical variations. Chemical complexity is rich in such fields because they contain three highly reactive interfaces—between water and rock, water and air, and rock and air. The fields also have different temperatures at different spots. Multiply all of this together: the wetting-drying cycles happening multiple times each day (think Old Faithful in Yellowstone), variable

THEO ALLOFS *Getty Images*

pool chemistries, highly reactive interfaces, the ability of pools to exchange compounds as geysers splash their contents back and forth, and an interconnected, fluid-filled, subterranean fracture network. When you do the math, it looks as if a terrestrial geothermal field of 100 springs can generate a million or more new combinations of conditions every year!

Each warm pond becomes an "innovation pool," a test bed in which adaptive combinations of molecules rapidly emerge and find ways to grow and reproduce or in which maladaptive combinations fall by the wayside, unable to keep up. It is likely that immense numbers of combinations might have been required to assemble the first primitive version of life, in which case the process would take hundreds of millions of years. But the numbers of combinations in terrestrial geothermal fields suggest that life could have originated and begun to evolve in as little as 10 million years, with the first stages beginning as soon as there was a stable crust peppered with volcanic landmasses amid the oceans, just more than four billion years ago.

VENTING DISAGREEMENT

NOT EVERYONE AGREES that surface hot springs are the most likely sites of life's beginnings. The deep-sea vent hypothesis is still alive and kicking. At NASA's Jet Propulsion Laboratory, biochemist Mike Russell has developed *Alvin*'s original discovery of hydrothermal vents into an alternative, elegant—but as yet unproven—model. In his scheme, mineral membranes that form minuscule pores within vent rocks initially separate alkaline water from more acidic ocean water. This produces a gradient of several pH units, similar to the difference between a solution of household ammonia and a glass of orange juice. The gradient is a form of energy that can be tapped; modern bacterial cells do exactly this to generate the ATP they need. There is another source of energy in the vents in the mixture of dissolved gases such as hydrogen and carbon dioxide. Russell and his colleagues have proposed that when carbon dioxide in ancient seawater mixed with hydrogen coming from the vents, the transfer of electrons from hydrogen to carbon dioxide could synthesize more complex organic compounds. In their view, the mineral compartments resemble cells, and the energy of pH gradients and hydrogen could ultimately evolve into a primitive metabolism required by the earliest forms of life.

The hot spring field and deep-sea vent hypotheses have some far-flung implications. Beyond guiding further explorations of life's beginnings on Earth, they point to different approaches to search for life on other planets and their moons. If the deep-sea vent origins theory is correct, the icy ocean worlds of Enceladus and Europa may be good places to look. On the other hand, if our model of fluctuating hot springs is right, then these worlds are unlikely to host life.

What about Mars? Although there is good evidence for shallow seas on Mars in the distant past, there are few signs of a global ocean or of tectonic spreading zones that create hydrothermal vents on Earth. If life depended on vents to begin, it was unlikely to have begun on the Red Planet. But if life on Earth originated in terrestrial hot springs, it could have also begun on Mars, which had the hot spring ingredients of widespread volcanism and water. Indeed, in 2008 the Spirit rover discovered 3.65-billion-year-old hot spring deposits in the Columbia Hills on Mars, about the same age as our Dresser hot springs, which did a great job of preserving early evidence for life on Earth.

Both the deep-sea vent and the land-based hot spring pools models have a long way to go before either can be deemed correct. The origin of life is like a jigsaw puzzle with many different pieces, and we do not know enough yet to put each one in the proper position. At the Dresser Formation, for instance, we do not understand what causes certain elements to become concentrated in different pools, how geothermal fields evolve over time, or how their different chemistries interact to synthesize or degrade organic molecules. We need to construct more sophisticated experiments of prebiotic chemistry in a series of warm little pools, studying how complex organic molecules form and how they interact and combine when encapsulated within membranes.

Both on land and in the sea, chemical and physical laws have provided a very useful frame around this particular puzzle, and the geologic and chemical discoveries described here fill in different areas. But before we can see a clear picture of the origin of life, many more pieces need to be put in place. What is exciting, however, is that now we can see a path forward to the solution.

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