

Ultramafics-Hydrothermalism-Hydrogenesis-HyperSLiME (UltraH³) linkage: a key insight into early microbial ecosystem in the Archean deep-sea hydrothermal systems

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Abstract. Since their discovery in the late 1970s, deep-sea hydrothermal systems have been considered as likely candidates for the origin and early evolution of life on Earth. However, while subsequent investigations have revealed a great diversity of modern deep-sea hydrothermal ecosystems, they have done little to shed light on the issues of the origin and early evolution of life, metabolism, cells, or communities. Phylogenetic, biochemical and geochemical clues all seem to point to the early evolution of hydrogenotrophic chemolithoautotrophy such as methanogenesis and sulfur-reduction, thus pinpointing the availability of hydrogen as one of the key elements needed for the early evolution of earthly life. Hydrogen-driven, photosynthesis-independent communities are very rare on the contemporary Earth, however, being unambiguously found only in subsurface environments of H₂-dominated hydrothermal systems. Such systems have been termed hyperthermophilic subsurface lithoautotrophic microbial ecosystems (HyperSLiMEs) (Takai *et al.*, 2004; Nealson *et al.*, 2005). The supply of abundant hydrogen and available inorganic carbon sources to power such communities is most likely coupled to hydrothermal serpentinization of ultramafic rocks and input of magmatic volatiles, both of which are related to specific geological settings. We propose here, on the basis of findings in the modern Earth and implications for the deep-sea hydrothermal systems in the Archean Earth, that “Ultramafics-Hydrothermalism-Hydrogenesis-HyperSLiME”, a linkage we refer to as Ultra H³, provided a suitable habitat for the early microbial ecosystem on the Archean Earth.

Key words: chemolithoautotrophy, deep-sea hydrothermal system, hydrogen, serpentinization, subsurface, ultramafic

Introduction

Discovery of deep-sea hydrothermal systems along Mid-Ocean Ridges (MOR) toward the end of the 1970s provided an expanded view of the deep sea, which had been long believed to be a cold, dark, and barren environment (Corliss *et al.*, 1979). Dense, abundant and diverse populations of animals were found associated with the active venting of superheated water, and it soon became apparent that these macrofaunal populations were strictly dependent on the primary production of symbiotic and free-living chemolithoautotrophic microorganisms, which obtain energy for carbon fixation, biosynthesis and other life activities from inorganic substances such as H₂S, CO₂,

H₂, CH₄ and so on entrained by hydrothermal fluids from the Earth's interior (Rau, 1981; Rau and Hedges, 1979; Jannasch and Mottl, 1985). Subsequent to the initial studies of the MOR systems, many other types of hydrothermal fluid geochemistry and hydrothermal fluid-associated ecosystem have been explored in the subduction zones, so-called Volcanic Arcs (VA) and Backarc Basins (BAB), of the Western Pacific region (Takai *et al.*, 2006).

The spectacular and unexpected properties of the deep-sea hydrothermal systems have attracted the interest of a diverse array of scientists, ranging from geophysicists (concerned with plate tectonics, mantle dynamics, magmatism, volcanology), to geochemists, microbiologists, and biologists. Not only are the sys-

tems a haven for extremophilic organisms, but they have inspired many ideas concerning their suitability as sites for the prebiotic chemistry required for the origin of life (Yanagawa and Kojima, 1985). Indeed, multiple lines of evidences from different fields (geology, chemistry, geochemistry, and molecular biology) have been used to argue that the deep-sea hydrothermal systems could have been the cradle of earthly life. For example, geologists have argued that the potentially most ancient microbial fossils are retrieved from a paleoenvironment that might be related to deep-sea hydrothermal systems in the early Archean (Rasmussen, 2000; Ueno *et al.*, 2001; 2004; 2006). Chemists have defined a number of reactions potentially associated with prebiotic chemical evolution (synthesis of amino acids, nucleotides and hydrocarbon, and polymerization of these molecules) that occur readily under the physical and chemical conditions characteristic of the deep-sea hydrothermal systems (Imai *et al.*, 1999; Yanagawa and Kojima, 1985).

In this paper, we discuss the possibility that irrespective of the origin of life, certain types of deep-sea hydrothermal systems might be havens for the types of communities that potentially represent the earliest microbial ecosystems. This discussion is somewhat associated with the last common ancestor concepts previously suggested as “progenote” (Woese, 1977; Woese *et al.*, 1990), “cenancestor” (Pace, 1991), “the last universal common ancestor (LUCA)” (Forterre, 1996) or “commonote” (Yamagishi *et al.*, 1991). Such thoughts derive for the most part from the interpretation of the molecular phylogeny of ribosomal RNA and other functional proteins, and lead to two contrasting notions of hyperthermophilic and mesophilic common ancestors, respectively. However, recent proteomic and protein engineering analyses have been used in support of the hypothesis for a hyperthermophilic common ancestor (Di Giulio, 2003; Iwabata *et al.*, 2005; Miyazaki *et al.*, 2001). Thus, it is reasonable for us to think that a certain high-temperature “window” occurred through which the ancient predecessors of today’s surviving life forms must have passed. The Archean deep-sea hydrothermal environment could be the window.

While we focus here on the early evolution of life and the potential characteristics of the ancient microbial community, it is also reasonable to point out that the diversity of hydrothermal environments might be an important factor by which to predict the nature of the microbial community inhabiting those environments. This is particularly true with regard to metabolic abilities—each type of hydrothermal system might offer different metabolic opportunities and pres-

ent different metabolic demands, leading to community formation of fundamentally different microbial metabolisms. With regard to the early microbial ecosystems in the Archean hydrothermal environments, one considers what kind of geological settings might host the ancient microbial ecosystems as predecessors of contemporary life. If one can come to some conclusion with regard to this question, then one can ask whether or not there are on the Earth today any environments that might serve as reasonable analogs to these ancient sites.

Here we propose a hypothesis that hydrogen-producing hydrothermal systems might favor the existence of the potentially earliest microbial communities. The fundamental ideas of this hypothesis are derived in part from recent results which suggest that there are metabolically dynamic communities existing in the subsurface that are geochemically isolated from any surface chemistry or biology, and that may serve as a good analogous model to the interrelationship between the Archean deep-sea hydrothermal environments and microbial ecosystems.

Hyperthermophilic subsurface lithoautotrophic microbial ecosystem (HyperSLiME) as a modern analog to the earliest microbial community

One approach to thinking about the hypothetical earliest microbial community is that of energy metabolism—what are the sources of energy available to different systems, and what kinds of communities or consortia might such systems have hosted? There are several different geochemical explanations for abundant energy sources in the Archean atmosphere and ocean (Habicht *et al.*, 2002; Kasting & Ono, 2006; Kharecha *et al.*, 2005; Tian *et al.*, 2005). Almost certainly, however, the major oxidant available to the emerging life was CO₂. Thus, early life forms were either fermentative, utilizing the limited amount of abiotically produced organic carbon and the inorganic fermentation sources of CO and S⁰ (no need for electron acceptors), and/or chemolithoautotrophic, utilizing the more abundant hydrogen (supposing that CO₂ is the primary electron acceptor). Self-sustaining communities would surely have needed to utilize inorganic energy and carbon sources which would have been abundant and accessible substrates provided by the hydrothermal activities. The onset of such hyperthermophilic chemolithoautotrophy could excrete biosynthesized organic molecules that could be utilized by heterotrophic fermenting organisms, thus leading to what we imagine as the earliest communities or ecosystems. It is also an important part of our hypoth-

esis that the nature of the hypothetical earliest ecosystem or community is not genomics-related, but metabolism-related: it must have been an assemblage of different metabolic abilities building to a self-sustaining community. To some extent it would be irrelevant whether these were the domain of Bacteria, Archaea, or Eucarya, despite the inability of contemporary Eucarya to grow chemolithoautotrophically.

The concept of the deep hot biosphere began with the speculations of Gold (1992), and the first such candidate for such a community called SLiME (sub-surface lithoautotrophic, microbial ecosystem) was proposed by Stevens and McKinley for a subsurface microbial ecosystem in the Columbia River Basalt system (1995), a claim that was hotly contested by Anderson *et al.* (1998). Recently, Nealson *et al.* (2005) discussed the definition and criteria of H₂-driven SLiME communities, and reviewed the evidence presented by several laboratories for and against the existence of SLiME communities. These authors also discussed the possibility that such SLiME communities might be adequate present-day analogs of ancient earthly ecosystems. To be brief, the criteria that must be met for a contemporary ecosystem are (i) that the energy driving the ecosystem should be of geological origin—this would include both the electron donors and electron acceptors (i.e., one thus imagines that a combination of H₂ and CO₂ should be the dominant and accessible redox couple for microbial chemolithoautotrophic metabolism), (ii) that neither electron donors nor electron acceptors produced by photosynthesis should be part of the ecosystem, (iii) that a community of appropriate organisms consistent with the proposed geologically driven metabolism should be present, and (iv) that this community should be shown to be active with regard to the metabolic hypothesis put forward for the ecosystem. An immediately apparent conclusion that seems to follow from these requirements is that specific subsurface environments are probably the only places in the modern Earth where it is operationally possible to escape the “pollution” of either the photosynthetically derived electron acceptors (O₂, NO₃ etc.), or the organic photosynthate (organic carbon) (Nealson *et al.*, 2005).

A number of H₂-driven SLiME communities have been recently proposed or considered: 1) subsurface groundwater environment in the Columbia River Basalts (Stevens and McKinley, 1995); 2) hot groundwater in the Lidy Hot Springs in Idaho (Chapelle *et al.*, 2002); 3) a seafloor environment of the Kairei hydrothermal field in the Central Indian Ridge (CIR) (Takai *et al.*, 2004a); and, 4) a seafloor environment of the Lost City hydrothermal field off the Mid-

Atlantic Ridge (MAR) (Kelley *et al.*, 2005). Of these, however, only the HyperSLiME found in the Kairei field meets the requirements laid down by Nealson *et al.* (2005) for a “true SLiME” community. This being said, however, some of the criteria were not met because measurements were not made, and there may, with some luck, be found in the future other “true-SLiME” communities.

Abundance of H₂ in the hydrothermal activity is a key indicator for the presence of a HyperSLiME community

All the deep-sea hydrothermal systems generate abundant geochemical energy, as demonstrated by the model in Fig. 1, but only rarely is the output dominated by hydrogen. This is important with regard to the “true SLiME” criteria denoted above, as some energy sources such as H₂S, while being excellent energy sources, are not able to form the biologically operative redox reaction with CO₂ or other potential oxidants provided via hydrothermal fluids. Recent studies of different deep-sea hydrothermal fields have revealed that such hydrogen-rich systems are rather rare (Table 1). The highest levels of hydrogen were seen for the Rainbow field and the Logatchev field of the Mid-Atlantic Ridge (MAR) (Table 1), which are ultramafic rock-associated deep-sea hydrothermal systems (Charlou *et al.*, 2002), and for the Kairei field (Table 1). The H₂ concentration of hydrothermal carbonate fluids in the Lost City hydrothermal field is highly variable but can also represent extraordinarily high concentrations of H₂ (Table 1).

Of the deep-sea hydrothermal fields in which the microbial communities have been investigated so far, strong evidence for a methanogenesis-dominated SLiME-like community was obtained only from the Kairei field (Kelley *et al.*, 2005; Nakagawa *et al.*, 2005a; 2005b; Shrenk *et al.*, 2004; Takai and Hori-koshi, 1999; Takai *et al.*, 2001; Takai *et al.*, 2003; Takai *et al.*, 2004a; Takai *et al.*, 2006). Kelley *et al.* (2005) have argued that the microbial communities in the carbonate structures of the Lost City field could be sustained by primary production via H₂-based energy-yielding metabolism. However, all the data from geochemical analyses of the fluids, stable isotopic characterizations of total inorganic carbon (TIC), total organic carbon (TOC) and lipid biomarkers, and molecular phylogenetic characterizations of the microbial communities (Kelley *et al.*, 2001; 2005; Shrenk *et al.*, 2004), suggest that the microbial communities in the carbonate structures are energized by either (or both) anaerobic methane oxidation (CH₄) or aceticlastic

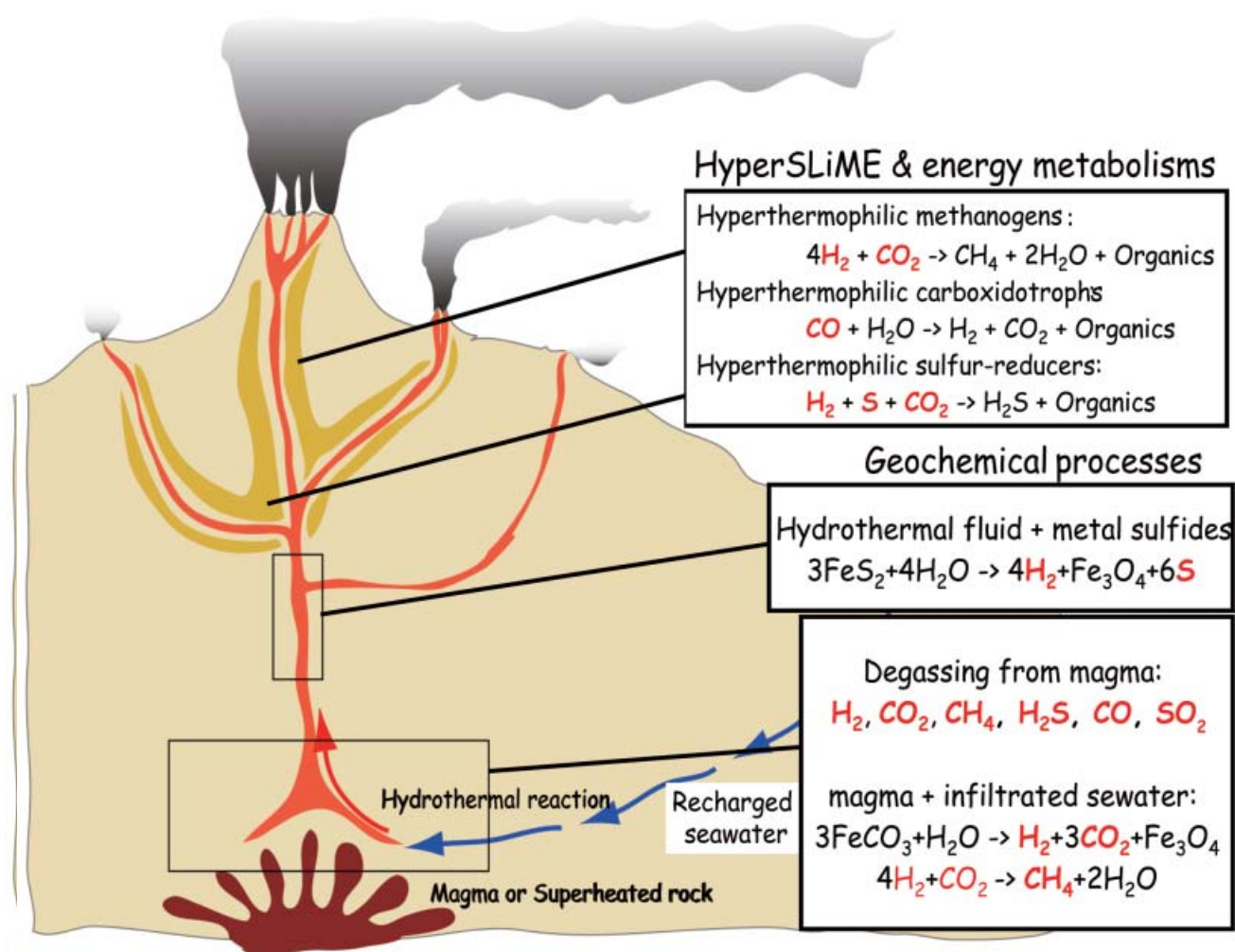


Figure 1. An original schematic model of a H_2 -driven, hyperthermophilic subsurface lithoautotrophic microbial ecosystem (HyperSLiME) sustained by H_2 from degassing of magma and hydrothermal reaction with metal sulfides before Takai *et al.* (2004). As described in the text, for HyperSLiME to prosper requires much more abundant H_2 than previously assumed. This leads to a new hypothesis, the “Ultramafics-Hydrothermalism-Hydrogenesis-HyperSLiME” linkage.

Table 1. Comparison of reaction rocks, H_2 concentration in hydrothermal fluids and presence of HyperSLiME or SLiME-like community among different deep-sea hydrothermal systems in the world.

Deep-sea hydrothermal field (location)	Reaction rocks	H_2 concentration (mM)	HyperSLiME or SLiME-like community	References for microbiology
Rainbow Field (on Mid Atlantic Ridge)	Peridotite & Gabbro	16	potentially (+)	Takai <i>et al.</i> , unpublished
Logatchev field (on Mid Atlantic Ridge)	Peridotite & Gabbro	12	n.d.*	
Kairei field (on Central Indian Ridge)	n.d.	2.5–8.5	(+)	Takai <i>et al.</i> , 2004
Lost City field (off Mid Atlantic Ridge)	Peridotite	< 1–15	(+) but strictly not SLiME	Kelly <i>et al.</i> , 2005 Schrenk <i>et al.</i> , 2004
TAG field (on Mid Atlantic Ridge)	Basalt	0.15–0.37	potentially (–)	Takai <i>et al.</i> , unpublished
Lucky Strike field (on Mid Atlantic Ridge)	Basalt	0.04–0.72	potentially (–)	Takai <i>et al.</i> , unpublished
Edmond field (on Central Indian Ridge)	Basalt	0.2	n.d.	
Suiyo Seamount field (on Izu-Bonin Arc)	Andesite	very low	(–)	Takai <i>et al.</i> , 2003
TOTO caldera field (on Mariana Arc)	Basalt & Andesite	very low	(–)	Nakagawa <i>et al.</i> , 2004b
EPR 21°N, 13°N & 9°N fields (on East Pacific Rise)	Basalt	0.1–1.8	n.d.	
Mariner Field (on Lau Basin)	Basalt	very low	potentially (–)	Takai <i>et al.</i> , unpublished
Izena hole field (on Okinawa Trough)	Andesite + sediments	0.05	potentially (–)	Nakagawa <i>et al.</i> , unpublished

*n.d., not determined. H_2 concentration data are based on Charlou *et al.* (2002), Gamo (1995), Kelley *et al.* (2005), Takai *et al.* (2004) and Van Dover *et al.* (2002)

methanogenesis (acetate) rather than by hydrogenotrophic methanogenesis (H₂). For the carbon source, the Lost City microbial ecosystem perhaps uses inorganic carbon in the seawater, rather than from the earth's interior (Kelley *et al.*, 2005). In addition, the molecular phylogenetic data suggest that the *Archaea*-dominated microbial communities in the Lost City carbonate structures are less thermophilic than HyperSLiME in the Kairei field. Thus, the Lost City microbial ecosystem does not represent a suitable analog for an early microbial ecosystem like HyperSLiME even though the occurrence of a H₂-driven chemolithoautotrophic community in the Lost City field may be still possible.

Other than the Kairei field and Lost City field, a potential H₂-driven SLiME-like community sustained by methanogenesis is not suggested because of the low levels of H₂ (Table 1). As Lovley *et al.* (1982) and Lovley and Goodwin (1988) have pointed out, methanogenesis exhibits a very high range of kinetic constants for H₂, and very high steady-state H₂ concentrations when compared to other anaerobic terminal electron-accepting reactions such as sulfate reduction, Fe(III) reduction, and Mn(IV) or nitrate reduction for growth. This implies that if H₂ concentration is low, hydrogenotrophic methanogenesis is prevented or outcompeted by other energy metabolisms. It thus seems likely that the occurrence and magnitude of HyperSLiME sustained by hyperthermophilic methanogens could be controlled by H₂ concentration in hydrothermal fluids of deep-sea hydrothermal systems. This builds up portions of the UltraH³ linkage hypothesis, a "Hydrothermalism-Hydrogenesis-HyperSLiME" linkage.

Mechanisms and geological settings for hydrogenesis in hydrothermal systems

Several major mechanisms of generation of H₂, i.e., hydrogenesis, in natural environments have been proposed: 1) degassing magma (Von Damm, 1995); 2) fault activity (Wakita *et al.*, 1980); 3) radiolysis of water (Savary and Pagel, 1997; Vovk, 1982); 4) microbial fermentation (Jackson and McInerney, 2002); and, 5) serpentinization (Janecky and Seyfried, 1986; Coveney *et al.*, 1987; Sleep, 2004). Although all these mechanisms may be to some extent involved in input of H₂ to hydrothermal fluids, the extraordinary abundance of H₂ in the hydrothermal fluids of the Rainbow, the Logatchev and the Lost City fields is explained by the serpentinization of oceanic peridotite, an ultramafic mantle rock (Table 1) (Charlou *et al.*, 2002; Kelley *et al.*, 2001).

Mantle peridotites and lower crustal plutonic rocks are significant components of the oceanic lithosphere. They are present in the shallow zones of ocean crust and are even exposed on the seafloor in many tectonic settings of the modern Earth (Früh-Green *et al.*, 2004). The alteration of these rocks, particularly alteration of the mineral olivine, by the process of serpentinization involves the oxidation of Fe(II) to Fe(III), and results in the production of heat through exothermic reactions, generation of reduced, highly alkaline fluids, and hydrogenesis, and of course the formation of the mineral serpentine (Figure 2). In reality, serpentinization involves a series of continuous metastable reactions governed by local variations in the activities of elements such as Si, Mg, Fe, Ca, C and H⁺ in the fluids (Früh-Green *et al.*, 2004). In addition, during the alteration of mantle peridotites and other ultramafic rocks, it is commonly believed that Fischer-Tropsch-type reactions have a great impact on the fluid chemistry associated with microbial energy metabolisms, that is, abiotic formation of CH₄ and other hydrocarbons consuming H₂ and CO₂ with FeNi alloys, magnetite and chromite as catalysts (Fig. 2) (Yoshida *et al.*, 1993; Berndt *et al.*, 1996; Foustoukos and Seyfried, 2004). A number of experimental investigations of seawater-ultramafic mineral reactions at high temperatures have demonstrated the formation of abundant H₂ and CH₄, and even detectable ethane, propane and formate (Janecky and Seyfried, 1986; Berndt *et al.*, 1996; Horita and Berndt, 1999). However, recent similar experiments have suggested that CH₄ might be produced from H₂ and reduced carbon sources (mainly graphite) in the minerals rather than aqueous CO₂ (McCollom and Seewald, 2001). In either case of the carbon source, Fischer-Tropsch-type reactions are able to deprive the post-serpentinization fluids of H₂ by abiotic methanogenesis, which may prevent the activity of a HyperSLiME or SLiME-like community based on microbial methanogenesis. In fact, the pore waters up to a depth of approx. 60 m below the seafloor of the serpentine mud volcano, South Chamorro Seamount, in the Mariana Forearc have no detectable H₂ despite high concentration of CH₄, and subsequent hydrocarbons are evidently produced from serpentinization-derived H₂ through Fischer-Tropsch-type reactions (Salisbury *et al.*, 2002; Mottile *et al.*, 2003). At all depths of core samples, no apparent molecular signature and culture for methanogenic *Archaea* has been identified (Mottile *et al.*, 2003; Takai *et al.*, 2005).

The equilibrium of abundance of H₂ and CH₄ formed throughout serpentinization hydrogenesis and Fischer-Tropsch-type reactions is strongly influenced

Forearc serpentine seamounts (Kelley *et al.*, 2005; Schrenk *et al.*, 2004; Takai *et al.*, 2005).

In contrast, the H₂-abundant hydrothermal fluids of the Rainbow and the Logatchev fields typically display a pH around 3. Such acidic hydrothermal fluids are common in deep-sea hydrothermal systems and are explained by input of magmatic volatiles, whether they are provided directly from magma itself or subsequently from the inclusion of magmatic volatiles in the heat-source rocks (Alt, 1995). In these deep-sea hydrothermal systems, therefore, the magmatic input might play a significant role in preparing the moderate pH conditions and the addition of inorganic carbon sources from the interior of the Earth. With the magmatic input, the microbial ecosystems in the ultramafic rock-associated deep-sea hydrothermal systems such as the Rainbow and the Logatchev fields might be energized by H₂ from hydrothermal serpentinization of ultramafics. In such geological settings, the complete linkage of UltraH³ (Ultramafics-Hydrothermalisms-Hydrogenesis-HyperSLiME) could be operative.

Searching for an UltraH³ site favorable to life on the modern Earth

So, we have drawn a picture in which UltraH³ creates a problem for itself via the formation of high pH conditions, and have proposed a solution for this dilemma, namely, that the problem might be solved by the mixing of low pH waters from magmatic systems, thus providing abundant H₂ at pH levels consistent with comfortable inorganic carbon chemistry. One such system may be the Rainbow hydrothermal field, which is located on a tilted ultramafic ridge in a non-transform offset of the AMAR segment on the Mid-Atlantic Ridge. The field, at water depths between 2270 and 2320 m, is based on a clear tectonic control by a network of faults generated both by the ridge and the non-transform system (Fouquet *et al.*, 1997; Fouquet *et al.*, 1998). The hydrothermal fluid chemistry reveals the considerable contribution of the hydrothermal serpentinization of peridotite (Charlou *et al.*, 2002), but the vigorous black smoker ventings and the rare earth elements (REE) composition of the fluids strongly suggests the considerable contribution of deep magmatic intrusion and the gabbroic dykes to the hydrothermal reaction (Fouquet, personal communication). It seems likely that the hydrothermal activity of the Rainbow field stands on the deep subsurface structure with a combination of host peridotite and intrusive gabbroic magma: potentially an ideal setting for the UltraH³ linkage. However, the search for a HyperSLiME-like community has been not yet

undertaken in the Rainbow hydrothermal field or other ultramafics-associated hydrothermal systems in the Mid-Atlantic Ridge.

The Kairei hydrothermal field is located in the first segment of the Central Indian Ridge (CIR-S1), which was the first deep-sea hydrothermal field discovered in the Indian Ocean (Hashimoto *et al.*, 2001). It is situated at the eastern axial valley wall very close to the inside corner of the ridge-transform intersection (RTI) between the first and second segments of the CIR (Fig. 3). The local bathymetric topography and the dive surveys using *DSRV Shinkai 6500* revealed that the hydrothermal activities of the Kairei field were distributed along the lava flow extending 100 m east to west and 40 m north to south. Based on the geochemical and stable isotopic characterizations of the hydrothermal fluids and the microbiological exploration of the seafloor microbial communities, the extremely high concentration of H₂ and significantly ¹³C-depleted CH₄ in the fluids, the possible occurrence of HyperSLiME was demonstrated, thus establishing the elements of the UltraH³ linkage (Hydrothermalism-Hydrogenesis-HyperSLiME) in the Kairei hydrothermal field (Takai *et al.*, 2004). What remains to be established is the contribution of ultramafic rocks to the hydrothermal reactions. This is strongly suggested by the extraordinary amount of H₂ in the fluids, but the association of ultramafic rocks with the hydrothermal activity has not been directly identified according to geophysical and petrological criteria.

Recent bathymetric and geophysical surveys of the southern CIR around the ridge-ridge-ridge type triple junction, the Rodriguez Triple Junction (RTJ), to which the Kairei field is closely situated, suggest that the ultramafic rocks should be relevant to the hydrothermal activity in the Kairei field. As a whole, the CIR has been characterized as an intermediate-rate spreading ridge, while the southern region of the CIR is based on variable morphological structures along the ridge axis, indicating spatial and temporal variation of magma supply (Honsho *et al.*, 1996). For instance, the inflated axial high at the southern end of the third segment of CIR (CIR-S3) seems to be a morphological signature observed in a fast-spreading ridge, but the deep axial valley of the first segment (CIR-S1) is similar to the axial valleys in typical slow-spreading ridges like the MAR (Fig. 3) (Briais, 1995). In such slow-spreading ridges with insufficient magma supplies, some significant portions of mantle peridotite reside within the shallow oceanic crust (Cannat, 1996) and very remarkable massifs characterized by corrugation perpendicular to the ridge axis, known as “meg-

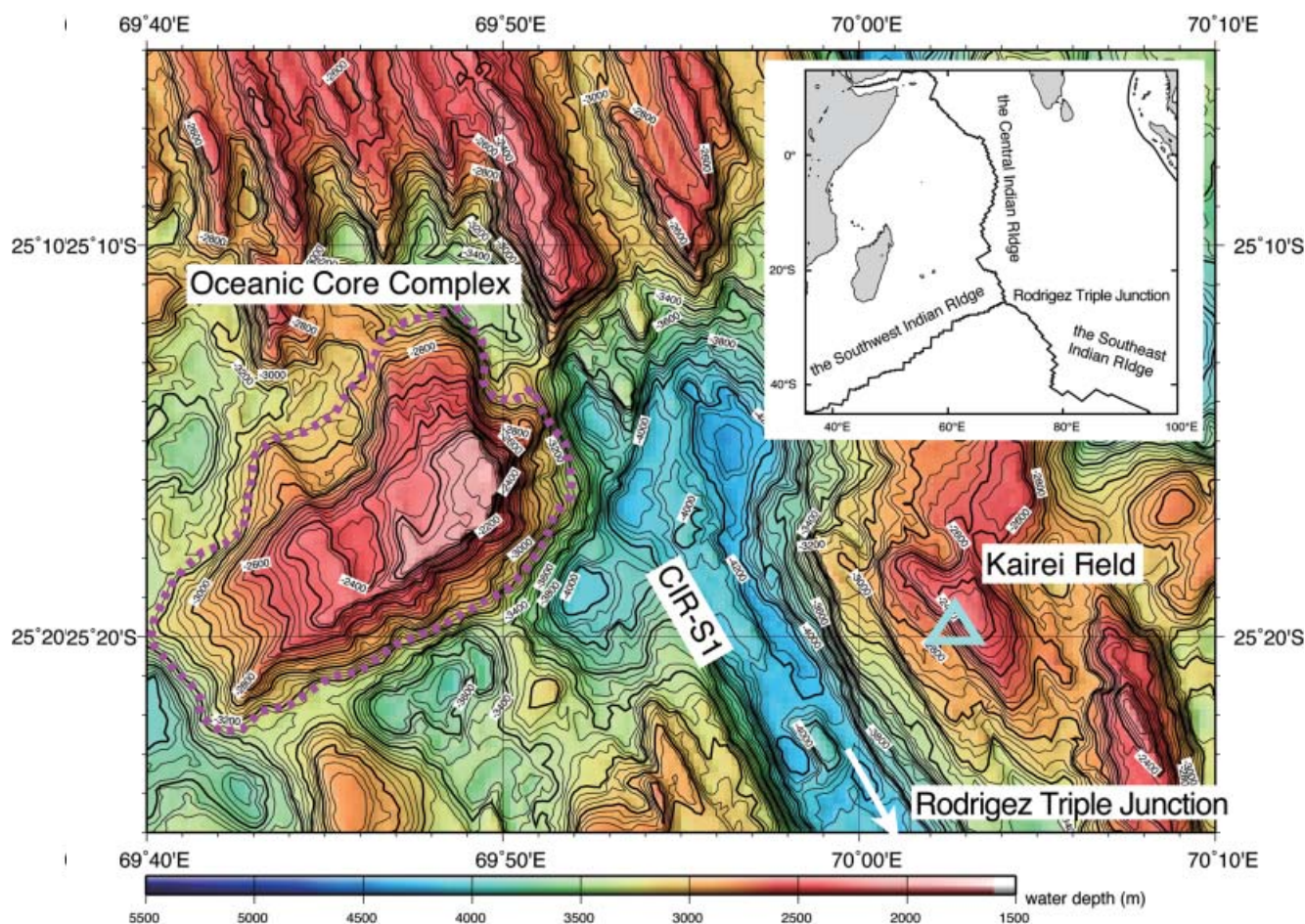


Figure 3. Bathymetric map of the area including the Kairei hydrothermal field and vicinity (modified from Kitazawa and Nakanishi, personal communication). Light blue triangle indicates the Kairei hydrothermal field. Dotted pink curve indicates Ocean Core Complexes (OCC)-like structure found.

amullions” or “Oceanic Core Complexes (OCC),” are frequently found (Cann *et al.*, 1997). This OCC-like structure is found on the western flank of the CIR-S1, at a position opposite the Kairei hydrothermal field (Fig. 3). The relevant area shows not only the typical morphological signature of OCC but also a strong positive gravity anomaly (i.e., Bouguer anomaly), indicating the existence of dense materials beneath the massif (Wakabayashi, 2003). From a volcanological aspect, the very short length of the CIR-S1 (< 50 km) might be consistent with a smaller amount of magma supply and a rapid cooling of magma by adjacent older lithosphere known as a transform effect (Klein and Lungmuir, 1987). In addition, the plate reconstruction approach of the RTJ and the southern CIR suggests that the plasticity of the RTJ and the very short lifetime of the CIR-S1 have been associated with an unstable setting of the RTJ (Honsho *et al.*,

1996; Mendel *et al.*, 2000). All these observations strongly suggest the geological setting of the CIR-S1 and the Kairei hydrothermal field is in fact associated with ultramafic rocks in the form of mantle peridotites, as inferred from the extraordinary H_2 concentration in the hydrothermal fluids. A series of geological, geophysical, and geochemical expeditions were conducted in early 2006 and the geological setting of the CIR-S1 and the Kairei hydrothermal field is now under extensive investigation.

UltraH³ linkage in the Archean Earth and implications for the early microbial ecosystem

The establishment of the UltraH³ linkage for the hydrothermal systems along slow-spreading MORs such as the Mid-Atlantic Ridge and the southern region of the Central Indian Ridge raises the question

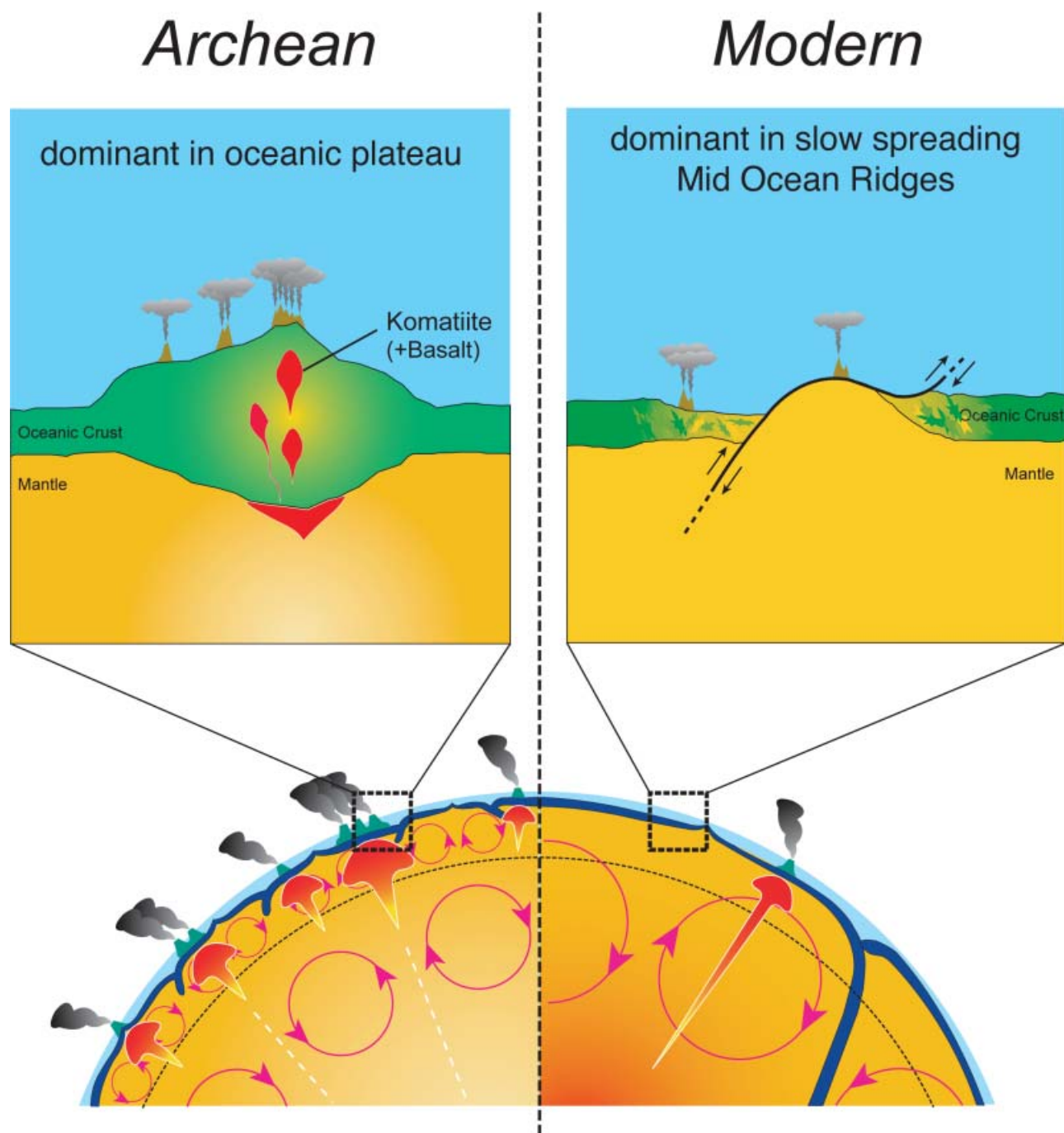


Figure 4. Schematic models of mantle convections, mantle plumes, plate tectonics and volcanisms hosting UltraH³ linkage in the Archean and modern Earth. In the Archean Earth, a hotter mantle might have induced smaller scales of and faster mantle convections than in the modern Earth, probably resulting in much more activity on MORs and a smaller scale of and more separated oceanic plates. Simultaneously, komatiite-dominated volcanism on ocean plateaus might have been abundant in the Archean Earth.

of whether similar UltraH³ linkages might have been continuously present over the long history of the Earth, and if so, whether they were important for the prosperity of early microbial ecosystems. That is,

could similar habitats have existed on the early Archean Earth ~3.5 billion years ago?

What might the Archean Earth have looked like with regard to ultramafic-associated hydrothermal sys-

tems and UltraH³ habitats? Key factors are chemistry and temperature of the Archean mantle, which presumably controlled the structure and chemical composition of the Archean ocean crust. It is generally assumed that ultramafic rocks (as well as mafic rocks) were more abundantly present in the Archean ocean crust than in the present day. Particularly, ultramafic lava flows known as komatiites have been discovered from almost all Archean cratons (Condie, 1994). Among the Archean supracrustal rocks termed greenstones, the volume fraction of komatiite and basalt is 40~80% (Condie, 1994), and some of the komatiite/basalt successions contain up to 25% of the komatiite component (de Wit and Hart, 1993). On the one hand, although dunites and harzburgites comparable to modern abyssal peridotite have been discovered in the early Archean gneiss complex (Friend *et al.*, 2002), mantle peridotite exposed onto the Archean ocean floor is assumed to be minor. The hotter Archean mantle probably conducted more extensive and active magmatism in the Archean MORs and formed a much thicker oceanic crust than in the modern Earth (McKenzie and Weiss, 1975; McKenzie and Bickle, 1988; Ohta *et al.*, 1996). Under such conditions, tectonically controlled exposure of mantle peridotite to the ocean floor would be very unlikely.

Komatiite is a distinctive volcanic rock of the Archean and is common in Archean greenstone successions, whereas it is unusual in the Proterozoic and quite rare in the Phanerozoic (Condie, 1997). The Archean komatiite and basalt flows are commonly pillowed (Condie, 2005), indicative of subaqueous eruption onto the seafloor. Komatiitic volcanism is considered to be involved in mantle plume activities, because of the very high estimated eruptive temperature of komatiite (Arndt, 1994). In addition, the voluminous oceanic eruptions of komatiite and associated basaltic rocks are commonly referred to as an oceanic plateau (Kusky and Kidd, 1992; Condie, 1994; Barley and Picard, 1999). Thus, komatiite might have been predominantly and ubiquitously distributed in the Archean ocean floor in the manner of the present day's oceanic plateaus.

The Archean submarine komatiites and basalts are often altered (e.g., albitized, silicified, carbonatized) by hydrothermal fluids in a similar manner to that seen in the basalts of the modern MORs and VAs (Barley, 1993; de Wit and Hart, 1993; Kato and Nakamura, 2003). The degree of hydrothermal alteration is significantly greater in the Archean oceanic komatiite/basalt than in the modern counterparts, probably due to greater heat and fluid fluxes in the Archean seafloor hydrothermal systems (de Wit *et al.*, 1987). Most of

the Archean komatiites were serpentinized by hydrothermal alteration immediately after eruption (de Wit *et al.*, 1987; Rollinson, 1999; Sproule *et al.*, 2002), and magnetite crystallization associated with serpentinization is also found in hydrothermally altered komatiites (Yoshihara and Hamano, 2004). Such petrological evidence strongly suggests that the seafloor hydrothermal serpentinization of komatiite which may have dominated in the Archean ocean floor could have been linked with abundant hydrogenesis. The komatiite-derived H₂, with CO₂ provided from the komatiite volcanism and the CO₂-rich Archean ocean (Grotzinger and Kasting, 1993), could have prepared widespread habitats suitable for early microbial ecosystems sustained by methanogenic primary production to prosper. In the Archean Earth, therefore, the UltraH³ linkage should be the komatiite-hosted version and it could have been much more abundant than the peridotite-hosted version in the modern Earth.

Concluding remarks

Here we have put forward arguments supporting an idea that we call the UltraH³ (Ultramafics-Hydrothermalism-Hydrogenesis-HyperSLiME) linkage hypothesis. This hypothesis claims: 1) that the ultramafic rock-associated hydrothermal system was the most plausible place for the existence of one of the earliest microbial ecosystems in the Archean Earth; 2) that H₂ generated through hydrothermal serpentinization could energetically support the activity of the early microbial community; and 3) that the H₂-abundant ultramafic-associated hydrothermal system has continuously harbored microbial ecosystems over the history of the Earth. The UltraH³ linkage is now being proven in the modern proxies situated in the MORs such as the Rainbow and the Kairei hydrothermal fields. However, direct exploration of the Archean version of the UltraH³ linkage based on komatiite is nearly impossible because of the lack of active komatiite volcanism in the modern Earth and significant differences in the ocean and atmosphere of the modern and Archean Earth. Indeed, it has been demonstrated that the mode of hydrothermal alteration in the Archean Earth should have been drastically affected by physical and chemical conditions of the Archean ocean and atmosphere (Kitajima *et al.*, 2001; Nakamura and Kato, 2004). Experimental work under conditions relevant to hydrothermal fluid-rock interactions and Archean conditions will be required before the physical and chemical conditions can be constrained.

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