

## ***Interactive comment on “Geodynamic and metabolic cycles in the Hadean” by M. J. Russell and N. T. Arndt***

**C. Blank (Referee)**

blank@wustl.edu

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I thought this was a very thought-provoking manuscript, because it makes an important attempt to tie the inorganic world with the organic; to identify potential early steps in the origin of life. This is a very difficult intellectual undertaking, given the lack of direct evidence, our poor understanding of the nature of the very early earth and where and when many of the important transitions may have occurred, our poor understanding of the spectrum of abiotic (and biotic) processes that may have been at work, and the interdisciplinary nature of the field.

Russel and Arndt make their case on how these transitions happened with firm, declarative statements which I believe, cannot be solidly made given the controversial and poorly understood nature of many of these statements. For example, their claim that the oldest zircons show the presence of an ocean by 4.4 Ga is one that is highly controversial where much more theoretical and experimental work needs to be done in order to explain the observations (e.g., Whitehouse & Kamber, 2002). I believe that

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the authors would have been better served if they would take their declarative statements, examine and explain the controversies, and then formulate their statements as hypotheses that need to be tested in order to support their own model regarding the transition between the inorganic and the organic worlds. When the controversies are not pointed out or discussed (granted this is difficult to do given space constraints), the declarative statements may appear one-sided.

I do not have sufficient expertise to make any specific comments about the first part of the manuscript and will leave this for others. However, I do have several comments on the declarative statements that the authors have made which are biological in nature:

1. The authors state (pg. 599; line 7) that "...because of the fragility of RNA we recognise that life is unlikely to have emerged at temperatures much above 40oC". This statement contains a number of underlying assumptions. First, it assumes that there was an RNA world (which is hypothetical and not universally accepted). Second, it requires that the RNA world, as proposed, must have existed at high temperatures or the RNA world must have existed at the same location as the origin of life. This is not necessarily the case as the two could have been separated spatially and/or temporally. Perhaps the origin of life did occur at high temperatures, and then moved into lower temperature environments once it reached the stage of the RNA world (if, in fact, the RNA world existed in the first place). On the other hand, there are many RNA molecules that have high G+C content and in their stems and therefore can maintain their secondary structures at high temperatures (even with several breaks in the sugar-phosphate backbone). With this information, one can then hypothesize that the RNA world indeed could have existed at moderately high temperatures if natural selection resulted in chemical and G+C content adaptations that conferred thermostability upon the molecules.

2. I do not understand the statement made (600;4) that ferredoxins are "proteins with the longest pedigree" (are the authors implying either that these proteins are more ancient than others in the cell, or that these proteins have the longest branch lengths

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and therefore fastest rate of evolution?). Is this a deduction based on the observation that iron and sulfur are thought to have been plentiful on the early earth and therefore it follows that proteins with Fe-S clusters (like ferredoxins as well as a large number of other redox proteins) are primitive? Or is this statement made because of the unique role that ferredoxins play in the cell (harvesting reducing equivalents at one location in the cell and then delivering them to wherever they are needed)? If the latter is the case, then I do not understand the point that the authors are trying to make. As far as I am aware, there is no phylogenetic evidence that these proteins are more ancient than any others that are currently found in the cell.

3. The authors state (600;25) "Indeed, acetate is the product of what is known as the acetyl coenzyme-A pathway - the most ancient for all metabolic pathways". I assume they mean the reductive acetyl Co-A pathway, which is reversible (and therefore could be used as an unusual form of respiration) and contains the enzyme carbon monoxide dehydrogenase (CODH). A quick BLAST search of sequenced genomes shows that the CODH enzyme is found in all the methanogen genomes (including Methanopyrus), as well as genomes from the Low G+C Gram Positive bacteria. Thus, one can infer that CODH (and the reverse acetyl Co-A pathway) was present in the ancestor to the Euryarchaeota, and then was later transferred laterally into the Low G+C Gram Positives. The enzyme is not in any of the Crenarchaeota or in any of the deeply branching bacteria, and therefore it is safe to conclude that this enzyme or this pathway was also not present in the last universal common ancestor (LUCA). So, I have to disagree with the statement made by the authors that this is the most primitive of metabolic pathways. There have been many papers in the last decade trying to infer the ancestral metabolic traits of the LUCA, and many of them contradict each other (some even claim it was totipotent, others not). Therefore, our picture of the nature of the LUCA's metabolic capability is somewhat murky at present and the claim of the authors premature.

4. The authors state (602;8) "Most prokaryotes near the root of the evolutionary tree can use Fe(III) as an electron acceptor, supporting the notion that this element was

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the first to fulfill this vital role". Statements similar to this are commonly seen in the microbiological literature. However, presence of a trait in the deep lineages does not necessarily mean that these traits are ancient, because these organisms have been around for a very long time and therefore contain many adaptive traits (such as adaptation to the presence of oxygen in the atmosphere, or adaptation to the presence of oxidized energy sources such as sulfate and thiosulfate) in addition to primitive traits. Therefore, in order to show that iron reduction is a primitive trait, the evolutionary history of iron reduction needs to be studied in detail in the deeply branching lineages. For starters, more thermophiles need to be tested for their ability to reduce iron (because iron reduction is not often tested during characterizations). The trait needs to be mapped onto the phylogenetic tree of life, the ancestral character states inferred, and the biochemical pathways of iron reduction in the deepest lineages need to be shown to be homologous before the statement can be made that iron reduction is ancestral and present in the root of the tree of life.

5. The authors need to be very careful about their terminology when they talk about methanogens and methanogenesis (603;12) in the context of pre LUCA metabolisms. The methanogens share a complex, highly evolved pathway with a large number of unique enzymes and co-factors. Most methanogens reduce CO<sub>2</sub> with H<sub>2</sub> completely to form methane, and this is likely the ancestral type of methanogenesis that existed in the ancestor to the Euryarchaeota. Methanogenesis using more complex organic compounds (e.g., formate, acetate, and alcohols) appeared only in late branches in the euryarchaeal tree. As such, methanogenesis using acetate (by the methanogenesis pathway) could not have been an ancestral metabolic energy source for the Euryarchaeota. However, one could hypothesize that some distant ancestor, long before the LUCA, could have used acetate as an energy source. However, the organism cannot be called a methanogen, or be equated with the modern form of methanogenesis.

6. With regard to differentiation into the bacterial and archaeal domains (603;20) - there is no particular reason (or evidence), that I am aware of, that they should have

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diversified in the same location and in the same location as the LUCA. The hypothesis that they diversified in two different locations is equally parsimonious at this point, due to lack of evidence supporting one of these hypotheses over the other.

7. I am not aware of any phylogenetic data showing that Mn reduction and sulfur reduction arose after iron and carbon dioxide reduction (603;27). In fact, it has long been argued (e.g., in Woese, 1987) that sulfur reduction is an ancestral trait in the tree of life. My own work supports this, and suggests that heterotrophy is just as ancient as autotrophy in the tree of life (and thus autotrophy is not necessarily more ancient). Also, there are at least 5 known ways that prokaryotes fix carbon dioxide, and thus this trait has arisen convergently a large number of times. This alone suggests that this is not an ancient trait, but one that is more derived (Blank, in prep.)

8. The authors state (604;1) that "We can say that Bacteria were initially suited to low to moderate temperatures, and the Archaea originally evolved to withstand the shock of relatively high temperatures (i.e. ~55oC). But the propensity to live well above 40oC was passed back to the nascent bacteria through genetic transfer." They the authors go on to explain that an impact could explain why the LUCA was a thermophile. These statements do not follow each other logically nor do they make sense, and the claims should be backed up with appropriate citations. Most would agree that the deepest branches in both the bacterial and archaeal domains live at very high temperatures, and therefore it is safe to infer that the LUCA as well as the ancestors that gave rise to the archaeal and bacterial domains were not just thermophiles, but hyperthermophiles. The French hypothesis and the work in Benner's lab (as indeed is correctly cited by the authors) are in the minority in claiming that the LUCA was not a hyperthermophile. Second, there are many instances where hyperthermophilic lineages independently diverged into low temperature environments. There are also instances where mesophilic ancestors diverged into high temperature environments (e.g., in the Cyanobacteria), however these secondary adaptations back to high temperatures never quite made it back into the realm of hyperthermophily. The reason why it is difficult/rare to go from

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low to very high growth temperatures is that a large number of mutational changes are needed. All the proteins in the cell need to adapt to higher temperatures so they don't denature. All the RNAs in the cell (rRNA, tRNA, other catalytic RNAs) have to adapt to remain folded at higher temperatures (e.g., by increasing the G+C content in their stems and loops). It is also likely that the cellular dynamics of co-factors and nucleotides have to be fine-tuned, since many of these are unstable at higher temperatures. The genome itself also has to be stabilized, either with supercoiling or with protective proteins, to eliminate thermal denaturation, double strand breaks, and chemical damage to the bases which can occur at more frequently higher temperatures. Hyperthermophiles also have some unique adaptations, like reverse gyrase, which help them survive at very high temperatures. Thus, it is doubtful that thermophily is a discrete trait that could be laterally transferred between distantly related lineages, as the authors propose. Lastly, yes it is agreed that one hypothesis to explain why the LUCA is a hyperthermophile is that it survived a meteorite impact. However, there are plenty of other hypotheses, which at this time should carry equal weight as the first in the face of a lack of evidence, such as that perhaps there were also mesophiles early on, but they were out-competed with organisms with hyperthermophilic ancestry. Or, the LUCA could have been seeded from another planet, such as Venus, Mars, or Europa, upon which the origin of life occurred (rather than the Earth).

9. It is not clear to me why the authors need to postulate some physical means (obduction of oceanic crust) in order to transfer microbes from the hydrothermal mounds on the oceanic floor to shallow water environments (605). The authors claim that their rationale for this hypothesis is that the ocean at the time was "inhospitable". It is not clear why the early oceans would have been inhospitable (or less hospitable than the hydrothermal mounds which are at the interface of a hydrothermal cell and the ocean). Perhaps they are referring to the near surface environments which are also part of the ocean but bombarded with intense uv? Perhaps the authors are implying that there would have been a general lack of energy sources in the early ocean? In general, microbes that are in stationary phase (where they are not actively growing) can exist

for very long periods of time. They also take on traits that render them resistant to environmental stresses like heat shock, cold shock, desiccation, as well as exposure to uv, ionizing radiation and peroxides. While in this state they are capable of acquiring mutations, so they can still evolve even if net population growth is not occurring. In this manner, cells released from their native environments (such as the mound) should be able to persist in a semi dormant, resistant state until they come in contact with growth conditions and energy sources they can survive in/on. For example, Thomas Brock showed several decades ago that one could culture thermophiles normally found in hot springs from samples of the Firehole River kilometers outside of Yellowstone National Park (despite their numbers being swamped out by simple dilution along with a vast number of actively growing low temperature biomass in the river). Thus, it is not immediately clear to me why an unusual event such as obduction of oceanic crust is needed to account for the transport of microbial cells from the deep ocean to shallow water environments on the early Earth.

10. The details of the evolutionary history of photosynthesis are poorly understood at this stage, despite decades of work by a large number of researchers. This is, no doubt, because that evolutionary history has been very complex, and was likely a combination of divergent as well as horizontal evolution with different sets of components having different evolutionary histories (see Olsen and Blankenship, 2004). Also, many of the genes in photosynthesis have fast evolutionary rates and therefore long branch lengths, so phylogenetic trees with these proteins are not easily resolved (requiring a large amount of data). At this time, it is not clear which photosystem came first (PS1 or PS2), particularly since it has been proposed that the Green Non-Sulfur bacteria, which branch somewhat deep in the bacterial domain and therefore has been long assumed to be primitively photosynthetic may have acquired photosynthesis laterally. It should be noted that PS2 is also found in the Proteobacteria and in the Green Non-Sulfur bacteria, and none of these groups are capable of splitting water to produce oxygen. Therefore, it is technically incorrect to say that "PS2 oxidizes water using what is known as the oxygen-evolving complex (OEC; 607;13)". Rather, what is unique is the

combination of the OEC and PS2 in the cyanobacteria which led to using water as the electron source for oxygenic photosynthesis which is the key evolutionary innovation.

11. The authors cite (608;9) three works as evidence for the early origin of photosynthesis. In the first citation, Dymek and Klein studied the REE composition of Isua (metamorphosed) banded iron formation. In their work, they state that "The Archean seawater component possessed a negative Ce anomaly, suggesting that ancient oceans may have been substantially more oxidized than was heretofore recognized". Nowhere in this paper is oxygenic photosynthesis implicated or implied as a culprit. Rather, this change in oxidation state could be due to a shift in Eh/pH space from the stability fields of reduced iron and cerium into the fields of oxidized iron and cerium. This does not necessitate the presence of oxygen or oxygenic photosynthesis (Dymek agrees with this opinion today, 16 years after this work was published; personal communication). A similar argument could be made for the second citation (Rosing and Frei). These authors show, interestingly, that high amounts of uranium were precipitated in early sediments, requiring an oxidizing environment so that uranium could be transported in solution and then delivered to the (reduced) site of deposition. Here too, this could be due to geochemical shifts in the Eh/pH of solutions, moving between the uraninite stability field and field of dissolved uranium ions. These shifts in solution chemistry do not necessarily require the presence of oxygen, and thus are not a definitive indication of oxygenic photosynthesis. Rosing and Frei, however, do have corroborating evidence of oxygenic photosynthesis in the form of a -25 per mil fractionation in organic carbon. Unfortunately, other autotrophic prokaryotes can fractionate carbon to a similar degree (House et al., 2003), as can abiotic processes (e.g., recent work of Tom McCollom). The third citation (Pace) is a conference abstract (enough said). Rather, the best evidence for the early presence of cyanobacteria is the hopanoid geolipids (Brocks et al., 1999; Summons et al., 1999; Brocks et al., 2003).

To conclude briefly: I believe that there are some important contributions in this work, such as the articulation of the hydrothermal mound as the cradle of life, what sort of

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inorganic and fledgling bio(in)organic processes could have occurred on the mound, and how organisms then moved from the mound to diverge into other environments with other phenotypes. Unfortunately, I have to disagree with a large number of the declarative statements that the authors make about evolutionary processes that would have occurred toward the later part of their scenario. Given that remark, it must be stated that there is a great deal to be learned in the early evolution of life on Earth, that this is a field seeing rapid progress, particularly given whole genome sequences, and that we should see a number of the controversies and problems better illuminated in the near future.

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