

July 16-21, 2017 at UC San Diego, CA, USA

Early Earth Environments for an Emerging RNA World – More Widespread than Previously Thought?

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The RNA World hypothesis offers a solution to the unlikely scenario for the co-evolution of DNA and proteins during the emergence of life. In order to realize an RNA World, however, there must be a polymerization pathway for abiotically synthesized, monomeric nucleotides to form the necessary, catalytic RNA oligomers. It is well established that select, activated montmorillonite clay samples can catalyze RNA oligomerization at ambient pressure, however, it is critically important to explore these and other pathways in the context of plausible, prebiotic geochemical and mineralogical environments on early Earth. In the present work, we broadened the search for oligomerization pathways to include not only a montmorillonite clay, but also nontronite, lizardite (a serpentinization product), and anorthite, which are likely to have been produced under the bulk chemical conditions of prebiotic Earth; sulfur-rich pyrrhotite and black smokers, which would have been abundant at the ocean floor; and calcite, which is a common mineral that was likely widespread on early Earth. The reactions were performed with imidazole-activated 5'-adenosine monophosphate under ambient, 5 kbar, and 10 kbar pressure. Matrix Assisted Laser Desorption Ionization-Time-of-Flight Mass Spectrometry (MALDI-TOF MS) was used to characterize the reaction products. Reactions using montmorillonite clay typically yielded linear oligomers of 10 or more nucleotides up to 5 kbar, and shorter oligomers at 10 kbar. Carbonates and carbonate-bearing phases, on the other hand, yielded longer oligomers as pressures increased. Nontronite, pyrrhotite and the black smoker chimney sample also yielded oligomer lengths that exceeded mineral-free control experiments, but the results did not vary with pressure. While the mechanisms of these reactions are not yet understood, the important discovery that co-varying mineralogy and experimental parameters such as pressure can lead to catalytic activity in samples previously thought to be non-catalytic suggests that geologically-relevant environments that could support an RNA World on early Earth may have been much more widespread than previously thought.