INTERACTIONS OF POLYMER BUILDING BLOCKS WITH FATTY ACID VESICLES IN LOW SALT SUPPORT THE FRESH-WATER ORIGIN HYPOTHESIS. Roy A. Black[1], Moshe T. Gordon[2], Caitlin Cornell[2], and Sarah L. Keller[2], [1]Dept. of Bioengineering, University of Washington, Seattle, Washington 98195 (royblack@comcast.net), [2]Dept. of Chemistry, University of Washington, Seattle, Washington 98195-1700

A major problem in understanding the origin of cells is explaining how the building blocks of (1) the membrane (most likely fatty acids in the first membranes [1]), (2) RNA (bases and ribose), and (3) protein (amino acids) were selected, concentrated and co-localized. We propose that fatty acid membranes, which assemble spontaneously in fresh water, bind the building blocks of the two polymers, and that this binding increases the formation and stability of the membranes [2]. The resulting selection, concentration and conformational constraints could have facilitated the formation of RNA and protein. We previously showed that RNA bases and ribose do bind to and stabilize vesicles composed of decanoic acid, a prebiotic fatty acid [3]. Here we present evidence that (A) SINGLE AMINO ACIDS and (B) DIPEPTIDES also bind to and stabilize such vesicles. We first established conditions under which the addition of these compounds does not change the pH of the decanoic acid solution, since pH alterations alone substantially affect fatty acid vesicle formation. These conditions include 100 mM NaCl. Results: (A) SINGLE AMINO ACIDS. We found that four prebiotic amino acids—alanine, glycine, serine and threonine—increase the formation of decanoic acid vesicles, as assessed by the turbidity of the solutions. To confirm that the increases in turbidity were due to more vesicles, we stained membranes with a fluorescent dye and then observed them by fluorescence microscopy. We found that these four amino acids increase both the number of vesicles and density of staining. The increase in membrane formation was not simply due to the increase in ionic strength caused by the amino acids, because four other amino acids—leucine, isoleucine, valine and proline—did not increase turbidity. The greater hydrophilicity of the set that does increase membrane formation suggests that the amino acid sidechains affect interaction with the vesicles. If amino acids interact with fatty acid membranes as these results suggest, then they might be expected to mitigate salt-induced flocculation of decanoic acid vesicles; our previous work showed that nucleobases and ribose reduce salt-induced flocculation and that the extent of reduction by the bases correlates with the strength of their binding. We found that two of the amino acids tested, leucine and isoleucine, do substantially reduce flocculation of decanoic acid vesicles by NaCl. Finally, we found that all eight amino acids bind to decanoic acid vesicles based on a filtration assay, and that the more hydrophobic ones bind more strongly than the others. (B) DIPEPTIDES. Several dipeptides also increased turbidity, including Ala-Ala, Ala-Gly, Ala-Thr, and Ala-Pro. As with single amino acids, fluorescence microscopy demonstrated that these dipeptides increase both the number of vesicles and density of membrane staining. Importantly, we found that Ala-Ala and Ala-Gly increase vesicle formation to a greater extent than their unjoined amino acids. Dipeptides that contained Leu, Ile, or Val did not increase vesicle formation, consistent with the lack of effect by these amino acids when they are unjoined. We confirmed that the dipeptides bind to the vesicles based on our filtration assay, and found that they tend to bind to a greater extent than unjoined amino acids. Regarding possible mechanisms of interaction, Ala-Ala-NH_2 and Pro-Ala did not increase turbidity, suggesting a free carboxyl group and a free amine are required. The co-localization of membranes with amino acids and peptides was an essential early step in the origin of cells, and the ability of fatty acid vesicles to recruit these compounds under low salt conditions therefore supports the fresh-water origin hypothesis.

References:

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