Incorporation of NH_3 into the proto-metabolism initiated from aldehydes supplied from the early atmosphere

*Xiaofeng Zang[1], Yuta Asakura[1], Kota Mouri[1], Yuichiro Ueno[1,2,3]

1. Department of Earth and Planetary Sciences, Institute of Science Tokyo, 2. Earth-Life Science Institute, 3. Japan Agency for Marine-Earth Science and Technology

In order to understand the origin of life, it is crucial to consider not only the synthesis of organic compounds, but also how these molecules are organized into sustainable chemical systems under plausible early Earth condition. In early atmosphere, photodissociation of CO_2 by solar ultraviolet (UV) produces CO, which could initiate the abiotic synthesis of precursor organics[1]. Formaldehyde (HCHO) has been known as the primary product from CO and water vapor in the atmosphere[2], though it is not the only product. Our recent studies demonstrated that 3 key aldehydes (HCHO, acetaldehyde CH_3CHO and glyoxal $(CHO)_2$) can be continuously supplied from CO_2, CO and H_2O in atmospheres[3][4]. Subsequently, aqueous reactions of these aldehydes can produce more than 100 species, including keto acids[4]. The self-organized reaction network from the small aldehydes seems to mimic biological metabolism, and thus is named as the Universal Metabolism Ancestor (UMA)[4]. Here, we report how UMA incorporate ammonia (NH_3), which could be available in the environment. NH_3 could be produced photochemically from nitrous oxide (N_2O) in a reducing atmosphere containing CO or H_2 without catalysts[3][5] and also available in the hydrosphere[6]. The results showed that when incorporating $NH_{3,}$ UMA system has the capability to produce (1) amino acids, (2) heterocyclic compounds such as hexamethylentetramine (HMT), imidazole, pyridine and their derivatives, (3) urea and (4) uracil. The production amino acids could be synthesized through either reductive amination or transamination of their precursor keto-acids, which are all detected in UMA system. Meanwhile, heterocyclic compounds could be produced from reactions of NH_3 with aldehydes, and some of these heterocyclic compounds have similar structure to coenzymes utilized in modern biological metabolism, thus could have catalytic feedback to UMA system. On the other hand, urea has been known to have kye roles in prebiotic chemistry, it can participate in the formation of pyrimidine nucleobases such as uracil[7], assist the formation of peptide bond[8] and phosphorylation of prebiotic molecules[9], which could be the next step of the evolution of UMA chemical system. The resulting abundance of detected amino acids is roughly consistent with thermodynamic expectation. Interestingly, the top abundant amino acids, which are proteinogenic, are all produced from the byproduct of UMA system, suggesting a potential selection mechanism.
Our results demonstrated that UMA’ s chemical system has capability to produce amino acids and heterocycles, and thus could transition into a higher dimensional system, namely UMA-N, which is more productive, potentially through catalytic feedback to the reaction network itself. This may provide a plausible mechanism to select molecules potentially leading to the emergence of life.

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