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Article / Book Information

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種別(和文)	論文要旨
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（博士課程）
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論 文 要 旨

THESIS SUMMARY

系・コース： Department of, Graduate major in	地球惑星科学　系 コース	申請学位（専攻分野）： Academic Degree Requested	博士 Doctor of　（　理学　）
学生氏名： Student's Name	藏　暁鳳	審査員主査： Chief Examiner	上野　雄一郎

要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

This thesis reports experimental study on abiotic synthesis of organic compounds from nitrous oxide(N_2O) under a reducing atmosphere. The discovery of this study reveals a new aspect of the role of N_2O in prebiotic chemistry on primitive Earth, and expand our current acknowledge on nitrogen cycle on primitive Earth as well as planetary atmospheric chemistry.

In Chapter 1, current knowledge about prebiotic synthesis of organic molecules is reviewed. Particularly, the synthesis experiments of amino acids reported to date are re-evaluated and the remaining problems are pointed out. Although HCN has been considered as an intermediate to synthesis amino acids through Strecker reaction and nucleobases on early Earth, its concentration and formation process of NH_3 in the atmosphere remain unknown. On the other hand, N_2O, which is produced from N_2 atmosphere by lightning discharges, is the second most abundant nitrogen compound in the atmosphere, and is expected to be an essential nitrogen source for prebiotic synthesis. However, abiotic sink of N_2O has been largely unknown. There are few experimental studies on N_2O, and its chemical processes in the reducing early terrestrial atmospheres are completely unknown especially. In this study, experiments on abiotic synthesis from N_2O were conducted, utilizing the two most abundant energy sources on primitive Earth - UV and spark discharge.

In Chapter 2, a series of UV-irradiation experiments are conducted. The results showed that NH_3 can be synthesized from N_2O under reducing atmospheric conditions such as CO or H_2. Furthermore, several amino acids can be synthesized from N_2O and CO even without metal catalysts. This work demonstrated, for the first time, that N_2O could be an important nitrogen source and provide a new process for synthesizing ammonia and organic nitrogen species, though the production mechanism of NH_3 has not yet been explained quantitatively.

In Chapter 3, the potential formation mechanisms of amino acids were investigated through further analysis. First, acid hydrolysis of the aqueous samples of UV experiment were conducted, and resulted 50 times more amino acids than free amino acids, indicating that amino acids precursors may be synthesized through the photochemical process. In addition, unhydrolyzed samples were measured and analyzed using Matrix-Assisted Laser Desorption/Ionization Mass Spectrometry (MALDI-TOF-MS) and High Performance Liquid Chromatography-Orbitrap Mass Spectrometry (HPLC-Orbitrap MS/MS). The results revealed that a variety of nitrogen containing heterocyclic compounds such as imidazole and hexamethylenetetetramine (HMT) were present in the sample. Among these identified heterocyclic compounds, HMT is known to produce amino acids after hydrolysis, indicating that these heterocyclic compounds are formed by UV irradiation to N_2O and become precursors. Furthermore, uracil was detected in trace amounts, suggesting that photochemical reactions in atmospheres including N_2O and CO are crucial synthetic processes, because they provide both amino acids and nucleobases simultaneously. Reaction between aldehydes and NH_3 could be an important process to form C-N bond during the process instead of HCN.

In Chapter 4, electric discharge experiments into a simulated atmosphere containing N_2O and CO were conducted. As a result, nitrite NO_2^- and nitrate NO_3^-, which were not produced by UV irradiation, are the main products. Although the discharge process also produced NH_3 and amino acids, the varieties and yields of the organic compounds from the reacted N_2O were much lower compared to UV irradiation. The results showed that N_2O and CO were not only the product species by lightning or corona discharge in atmosphere, but they also have the potential to generate NH_3 as well as amino acids at the same time by corona discharge. However, the main pathway for amino acids synthesis from N_2O is atmospheric chemical reaction by solar UV light.

In Chapter 5, the supply flux of NH_3 and glycine to the surface from atmospheric N_2O were estimated according to the experimental results from Chapter 2~4. Two models are conducted, and photochemical supply of free glycine from the atmosphere is calculated to be 4×10^7 kg/year, assuming an early Earth atmosphere (CO_2: 0.1%, CO: 1%, N_2O: 3ppm) composed mainly of N_2 (1 bar). The value could reach to 10^8 kg yr^{-1} after fully hydrolyzed. This flux is comparable to the previously estimated production rate of glycine from lightning discharge in an N_2-CH_4 atmosphere or from cosmic ray irradiation of an N_2-CO(50%) atmosphere. Therefore, the photochemical process of N_2O in a reduced atmosphere may have been the primary pathway for amino acid supply processes on the early Earth before the arose of the first life.

備考 ： 論文要旨は、和文 2000 字と英文 300 語を 1 部ずつ提出するか、もしくは英文 800 語を 1 部提出してください。
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