論文の内容の要旨

論文題目

Organic aerosol production in reducing atmospheres and its influence on the radiation fields on Titan and early Earth（還元的大気における有機物エアロゾルの生成とタイタンおよび原始地球における放射伝達過程への影響）

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Organic aerosols are considered to be produced in CH_4-containing, reducing atmospheres, such as Titan, early Earth, and exoplanets. Physical processes (radiative transfer and microphysical processes of the organic aerosols) and chemical processes (photochemical and ion reactions to form organic aerosols) in these atmospheres are mutually dependent on each other. Thus, self-consistent coupling of the physical and chemical processes is essential to understand atmospheric and environmental evolution of these planets and satellites. However, many of the previous studies discuss the physical and chemical processes independently, and there are only a few studies that couple these two processes.

One of the large obstacles that prevent the coupling between the physical and chemical processes is uncertainty of production mechanism of organic aerosols in reducing planetary atmospheres. To filling the gap between the chemical and physical processes, we conduct laboratory experiments of formation of organic aerosol analogues, called tholin, and gas analysis of intermediate molecules formed by photochemical reactions using a hydrogen/helium lamp. We also perform photochemical calculations that simulate reactions in the laboratory experiments to identify the parent molecules and corresponding reactions that control the tholin production in the experiments. We measure the dependences of tholin production rate on both actinic UV flux and CH_4/CO_2 ratio. Our experimental results show that the tholin production rate is a linear function of UV flux, which suggests that the aerosol production is limited by polymerization reactions between intermediate products, i.e., parent molecules, produced from the photochemistry of CH_4. We also found that the aerosol production rate remains almost constant for a wide range of CH_4/CO_2 ratio when the CH_4/CO_2 ratio is in excess of unity. On the other hand, the tholin production rate decreases dramatically with a decline in CH_4/CO_2 ratio when the CH_4/CO_2 ratio becomes less than unity. Our photochemical calculations show that this behavior of tholin production rate is in a good agreement with polymerization reaction rates involving aromatic hydrocarbons, benzene. These results suggest that benzene is the parent molecule that controls the tholin production. On the other hand, polymerization reactions involving polyynes do not explain the measured tholin production as a function of CH_4/CO_2 ratio. Our results are considered to be applicable to estimate the production of monomers in a CH_4-contaitning reducing atmosphere using a one-dimensional photochemical model, which is necessary to couple the chemical processes with the physical processes in the atmospheres.

Based on our experimental results, we calculate the atmospheric composition, organic aerosol production, and its optical depth for variable planetary atmospheres, using a coupled

photochemical-microphysical-radiative transfer models. In Titan's atmosphere, the primary energy source for organic aerosol formation has been unclear. Our results show that polymerization reactions of benzene induced by solar far UV light cannot explain the proposed aerosol production rates in Titan's atmosphere. In contrast, our results suggest that the organic aerosols in Titan's atmosphere are largely produced by nitrile polymerization, which is initialized by irradiations of high-energy particles. There results suggest that the irradiation of high-energy particles is the primary energy source for the formation of thick aerosol layers in Titan's atmosphere. These conclusions further imply that Titan's climate and surface environments would have been very sensitive to variations in the magnetic field of Saturn and solar wind flux.

In early Earth's atmosphere, organic aerosols formed from atmospheric CH_4 are suggested to play key roles in determining the surface temperature. If their UV optical depth were thick, the organic aerosols would have possessed indirect greenhouse effect, which shields strong greenhouse effect gas NH_3 from UV light. On the other hand, if their optical depth in visible wavelength were thick, they in turn would have had anti-greenhouse effect, which cools the surface. Whether the organic aerosols work as indirect greenhouse or anti-greenhouse critically depends on their production rate in the atmosphere. Our results show that the organic aerosol layers produced by benzene polymerization induced by solar UV would have been optically thin, insufficient to have both indirect greenhouse and anti-greenhouse effects. The absence of strong anti-greenhouse effect in turn suggests that the greenhouse effect of ethane would have worked efficiently to keep early Earth's surface above freezing point of H_2O, especially under CH_4-rich atmospheric conditions. The supply and loss of both CO_2 and CH_4 through biogeochemical processes vary with surface temperature. Given that ethane concentration also changes with CH_4/CO_2 ratio of the atmosphere, there would have been a feedback relationship between surface temperature and ethane concentration. On the other hand, we found that thick aerosol layers would have been formed if aerosol production also proceeds through nitrile polymerization, induced by irradiation of high-energy particles. Such thick aerosol layers are optically thick sufficient for shielding NH_3 from UV light on early Earth. Further investigation of nitrile polymerization by laboratory experiments will be important in future studies to evaluate the role of organic aerosols in early Earth's atmosphere.