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Organic aerosol experiments for CH_4/CO_2 atmospheres using a hydrogen/helium UV lamp

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Organic aerosols are photochemically produced in CH_4-rich reducing atmospheres, but their production mechanisms are not well constrained. Organic aerosol layers are believed to influence the surface temperature of early Earth, through its anti-greenhouse (Pavlov et al., 2001) and/or indirect greenhouse effects (Wolf and Toon, 2010), however, because of the uncertainty of the aerosol production mechanism, there are large uncertainties inherent in previous estimates of the aerosol production rate and optical depth of aerosol layers (Trainer et al., 2006). In order to put a constraint to the production mechanism and obtain aerosol production rate applicable to CH_4/CO_2 atmospheres, we conducted laboratory experiments to form organic aerosol analogues using a hydrogen/helium lamp that simulates solar far UV (FUV) with wavelengths longer than 110 nm. We measured the aerosol production rate as functions of UV flux and of CH_4/CO_2 ratio in the reactant gas. The aerosol production rates were determined by ellipsometrically measuring the growth rates of thin organic films deposited on a substrate. The UV fluxes from the hydrogen/helium lamp were measured by N_2O/CO_2 actinometry. Our experimental results show that the aerosol production rate is not a second-order function but a linear function of UV flux. This leads to a lower estimate for aerosol production rate due to FUV irradiation, when extrapolating the production rate in Titan's atmosphere to early Earth and exoplanets. We also found that the aerosol production exhibits a steep decrease when the CH_4/CO_2 ratio becomes less than unity. In order to interpret the dependence of aerosol production rate on the CH_4/CO_2 ratio, we also performed one-box photochemical calculations, including 791 reactions and 134 species up to C_8 hydrocarbons. The one-box photochemical model was validated against some basic carbon species (CH_4, C_2H_2, C_2H_4, C_2H_6, CO, CO_2), in which the abundances of those species calculated with the model and observed with a quadruple mass spectrometer (QMS) show a good agreement. We found that the observed production rate is in a good agreement with polymerization reaction rates involving aromatic hydrocarbons (i.e., benzene), suggesting benzene is the key parent molecule controlling the aerosol production. On the other hand, polymerization reactions involving polyynes do not account for the experimental data, suggesting that they are not the limiting molecules. This implies that aerosol production rate in an early Earth atmosphere due to solar FUV would become significantly lower than a previous estimate which includes polymerizations of polyynes as formation reactions of aerosols (Pavlov et al., 2001), resulting in an optically thinner aerosol layer by a factor of 100. Thus the optical depth of organic aerosol layers produced by solar FUV in an early Earth atmosphere would not have had efficient anti-greenhouse effect or indirect greenhouse effect, which makes other greenhouse effect important for the Archean climate, such as greenhouse effect of ethane. We will also discuss the possibility of aerosol formation through nitrile reactions driven by high-energy particle irradiation, which could be more efficient than the aerosol production due to solar FUV.

Keywords: organic aerosol, photochemistry, laboratory experiment, reducing atmosphere