Geophysical Research Abstracts
Vol. 15, EGU2013-6544, 2013
EGU General Assembly 2013

Production of nitrogen oxides by lightning in a methane-rich early atmosphere

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The composition of the early Earth's atmosphere is not known. Assuming that rapid weathering of fragments from impacts took placed and efficient sequestration of carbon occurred in the Earth's mantle, the early atmosphere would have been mostly composed of molecular nitrogen with low concentrations of carbon dioxide (less than percent). In order preserve the existence of oceans, it is required to warm up the atmosphere almost exclusively with methane [1]. Predicted methane concentrations in the distant past range from few ppm to several thousand ppm. Photochemical models predict a production rate of hydrogen cyanide of the order of 6 Tg/yr in a 3 percent carbon dioxide atmosphere with 1000 ppm of methane [2]. When the atmospheric levels of carbon dioxide dropped to 0.3 percent but with the methane levels of 1000 ppm, the production rate of hydrogen cyanide increased up to 20 Tg/yr [2]. The nitrogen fixation rate by lightning in atmospheres dominated bymolecula nitrogen, less than 10 percent carbon dioxide, and the absence of methane has been reported by Navarro-Gonzalez et al. [3]. Here we report an experimental study of the effects of lightning discharges on the nitrogen fixation rate during the evolution of the Earth's early atmosphere from 10 to 0.5percent of carbon dioxide with methane concentrations from 0 to 10,000 ppm in molecular nitrogen. Our results show that the main nitrogen fixation products by lightning are nitric oxide, nitrous oxide and methyl nitrite. Preliminary estimates indicate that the production of nitric oxide is not dependent on the initial concentration of methane and that its production rate decreases from about 0.02 Tg/yr to about 0.003 Tg/yr in atmospheres ranging from 10 to 0.5 percent of carbon dioxide, respectively. Nitrous oxide is produced by lightning is the contemporaneous oxygenated Earth's atmosphere [4], but has not been detected in nitrogen-carbon dioxide mixtures in the absence of oxygen [5]. This is the first report for the production of methylnitrate by lightning in the early atmosphere. Further analyses are underway to accurately determine the production rates of these nitrogen oxides by lightning, and to understand their role for chemical evolution and early Earth's climate.

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