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Speciation and isotope ratios of nitrogen dissolved in the primitive ocean.

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Reconstruction of the geochemical cycle of N in the Early Archean is important to understand the origin and evolution of life on the Earth. Therefore, it is important to determine speciation and isotope ratios of inorganic N in the primitive-ocean and atmosphere from geological records.

We first report the speciation and isotope ratios of N in fluid inclusions preserved in 3.5 Ga hydrothermal deposits (silica dikes and quartz veins) from the North Pole area of the Pilbara Craton, Western Australia. Crush-leach analysis and Raman microspectrometry revealed that N within the fluid inclusions exists as N2 and NH4+. A negative correlation between the SO42/Na and 40Ar/36Ar ratios of the fluid inclusions suggests mixing of two end-members; hydrothermal fluid with low SO42-/Na+ and high 40Ar/36Ar ratios, and 3.5 Ga seawater with high SO42/Na and low 40Ar/36Ar ratios. Values of d15NN2 from the hydrothermal component vary over a considerable range (-3.0 ˜+3.7permi;), and those of the seawater component are well within this range (i.e., -0.7 ˜-0.2permil). This suggests that the isotope ratio of N2 dissolved in the 3.5 Ga seawater would have been -0.7 ˜-0.2permil. Since isotope fractionation between N2 in the atmosphere and N2 dissolved in seawater is minimal, the d15NN2 value of the 3.5 Ga atmosphere would have been within the range -2 ˜0permil, which is similar to the d15NN2 value of the present-day atmosphere (d15NN2 = 0permil). This study also suggests that the fluid inclusions contain NH4+ that would have been derived from the seawater and/or mantle at 3.5 Ga. Therefore, kerogens in Archean cherts might have been partly derived from biological assimilation of NH4+ in hydrothermal fluids. Also, isotope ratios of NH4+ would constrain the production process(es) of NH4+ on the Early Earth.