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Search for abiotic N_2 reduction in hydrothermal systems

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Abiotic N_2 reduction (ANR) has been considered as one of the fundamental first-step reactions for the origin of life and could have played a key role in the geological nitrogen cycle on the early Earth, but surprisingly poorly constrained. The difficulty in identifying ANR in the field is the lack of robust tools to distinguish the ANR-produced ammonium from the overwhelming biologically processed ammonium. Here, we analyzed the nitrogen isotope behavior during ANR. When equilibrium isotope fractionation dominates, the ANR-produced ammonium is ~13‰ (70 °C) to ~2‰ (800 °C) higher than the N_2 reactant [1]. This yields a $\delta^{15}N$ range of -3‰ to +13‰ for the ANR-produced ammonium, which is indistinguishable to the organic $\delta^{15}N$ range of -4‰ to +15‰. However, increasing experimental and field studies show that abiotic reactions involving N_2 (either as reactant or product) are more likely dominated by kinetic isotope fractionations. Based on available data from laboratory experiments and theoretical calculations, the kinetic isotope enrichment factors ($_{NH4+–N2}$) are determined to be –16‰ to –13‰ over 600–800 °C [2]. The $_{NH4+–N2}$ values at lower temperatures cannot be quantified yet due to the lack of data, but are expected to show even greater magnitudes. Consequently, the kinetic isotope effect would give the ANR-produced ammonium abnormally low $\delta^{15}N$ values (e.g., < –10‰) that are distinct to those of known Earth's reservoirs. Therefore, abnormally low $\delta^{15}N$ values of ammonium in minerals and fluids may serve as a sensitive tracer for ANR. In field studies, abnormally low $\delta^{15}N$ values have been reported from a variety of geological settings, including altered oceanic basalts in the Pacific Ocean [3], Atlantic Ocean [4], and South China Sea [2], as well as metamorphosed basalts in ancient subduction zones [5,6]. This suggests that ANR could occur widely on spatial and temporal scales.

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