Search for primordial biogenic isotopic signatures of nitrogen in Archean sedimentary rocks

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A significant portion of nitrogen in sedimentary rocks is considered to have a biological origin, trapped in organic forms, or as ammonium ions which often substitute potassium in minerals. Compared to other biomarkers such as C or S, N is characterised by its least chemical activity and its highest volatility. The former characteristics, largely owing to the exceeding stability of N₂ molecules, which strictly limits inorganic transmission of N from the atmosphere to the biosphere, may lead to construction of a simpler biological flux model of N compared to other elements. The latter characteristics suggest that N isotopic information in old sedimentary rocks is fragile, potentially disturbed by various later geological events, thus careful evaluation of the data is required. We consider that separation of N in different sites of rock samples and identification of their trapping forms are important first steps for such evaluation.

We analysed volatile compositions (isotopic compositions of N and Ar, and concentrations of C and H₂O, extracted using a stepwise combustion method) of an Archean chert representing a silicified rhyodacitic pyroclastic sediment from Kitty's Gap, Western-Australia, with a U-Pb age of 3.5 Ga. From the chert, composed of alternating black and white layers, 7 layers with different K₂O contents between 0.13 to 1.7 wt% were separated. Followed by analyses of bulk samples from respective layers, we further analysed K,Al-bearing phyllosilicate (hydromuscovites, HM) and quartz separated from each layer. A clear correlation between N and K or radiogenic ⁴⁰År concentrations among the bulk and separated samples suggests that bulk of N seems to be concentrated in HM. Among the bulk and HM, an interesting two-components mixing trend was observed in the δ^{15} N-⁴⁰Ar*/N diagram, suggesting existence of two N components with $\delta^{15}N$ values of <0 and +8 permil. The former N component, highly correlated with radiogenic ⁴⁰Ar, is probably trapped as NH₄⁺ in the K-site of HM, whereas the latter component, apparently concentrated also in the HM, seems to be trapped in a different form. The relationship of the two N components with clearly different isotopic ratios, possibly coexisting in the same phyllosilicate, will be discussed.