Goldschmidt 2021 Abstract
https://doi.org/10.7185/gold2021.6156

Nitrogen isotope variations across the 3.4 Gyr Buck Reef Chert, South Africa, question early nitrogen sources and pathways

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Nitrogen is a key element of the biosphere, present in numerous macromolecules and involved in major metabolic pathways. Its isotopic composition is controlled by metabolic activity and redox speciation. Reconstructing the biogeochemical nitrogen cycle on the Early Earth is therefore valuable to our understanding of the evolution of the atmosphere, ocean and biosphere. The present study intends to fill in the sparse Paleoarchean record by providing N isotope data from the ICDP BARB3 drill core [1] through the Buck Reef Chert sedimentary unit (3416 Myr), Barberton Greenstone Belt, South Africa. The nitrogen concentration and isotopic composition of 33 samples have been analyzed by dual-inlet mass spectrometry after sealed-tube Dumas combustion, extraction and purification. The exceptional preservation of organic matter [2] underlined by high TOC contents (up to 6%), low TOC/TN ratios (<145) and reported occurrences of biofilms [3] makes the Buck Reef Chert a perfect candidate to explore early biological cycle of nitrogen. $d^{15}N$ values range from -0.7‰ to +5.1‰, with marked positive values (mean $d^{15}N$ = +2.7‰) in the lower silicified part of the core, interpreted as shallow platform facies [3], followed by a clear stratigraphic decrease from +5.1‰ to -0.5‰ as we dive deeper towards the more basinal, siderite-rich, banded iron formation facies. Although various redox species of nitrogen associated with a complex cycle at a biofilm-scale cannot be excluded, the isotopic variations displayed by $d^{15}N$ values seem consistent with the range of fractionation associated with biological fixation of N_2 by diazotrophs using alternative nitrogenases. Indeed, lower $d^{15}N$ values might be consistent with an increasing contribution of (Fe)-nitrogenase in sideritic facies, where Fe is in excess compared to Mo. Alternatively, this stratigraphic $d^{15}N$ evolution could result from a mixing between a ^{15}N-enriched continental source (+2-3‰) of fixed nitrogen and hydrothermally ^{15}N-depleted ammonium (-5‰). Finally, this study suggests that different nitrogen sources or fixation pathways sustained a flourishing early Earth biomass as far back as 3.4 Gyr.

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