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Constraining the origin and prevalence of biological N2 fixation in the Precambrian

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Nitrogen is an essential nutrient for all life on Earth; however, atmospheric N2, the largest nitrogen reservoir at the Earth's surface, is chemically inert and only accessible to some prokaryotic microbes that possess a nitrogenase enzyme. Prior to the origin of this metabolism, bioavailable nitrogen may have been derived from hydrothermal activity, lightning or photochemical reactions, but these sources are minor today and probably became limiting with the expansion of the biosphere. The origin of biological N2 fixation was therefore of paramount importance for early evolution. Geochemical and phylogenetic data, however, suggest that this event may have been delayed until the early Proterozoic, possibly by the lack of dissolved Mo, an essential co-factor in nitrogenase. Here we show new nitrogen isotopic data from low-grade sedimentary rocks of the Soanesville Group (∼3.2 Ga, Western Australia), the Witwatersrand Supergroup (∼2.9 Ga, South Africa), and the lower Fortescue Group (∼2.75 Ga, Western Australia), with a total mean of 0.0 ± 1.2 ‰ relative to atmospheric N2. These values are inconsistent with abiotic sources of fixed nitrogen and difficult to reconcile with alternative nitrogenase enzymes that do not depend on Mo. Instead it is most likely that Mo-based nitrogenase had already evolved and was widespread in the mid-Archean. Combined with a literature database of δ15N values through time, our results suggest that other forms of fixed nitrogen became available in the late Archean and persisted throughout the Paleoproterozoic. In the Mesoproterozoic ocean, fixed nitrogen was likely restricted to shallow waters, while offshore environments were dominated by Mo-based N2 fixation. This basinal gradient likely disappeared in the Neoproterozoic. In conclusion, biological N2 fixation is a much more ancient metabolism than previously proposed and Mo has been bioavailable in at least small amounts throughout the Precambrian.