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Igneous rocks as a viable source of fixed nitrogen to the prebiotic world

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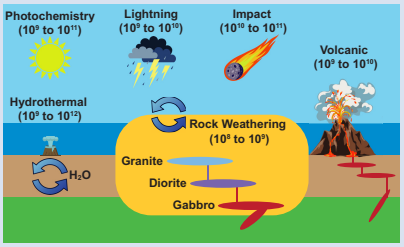
Abstract



The origin and early evolution of life on Earth and other habitable worlds requires constant supply of ammonic nitrogen (N). Previously proposed abiotic ammonium sources rely on sporadic and heterogeneously distributed high energy processes, such as lightning, subaerial volcanic degassing, or deep sea hydrothermal vents to generate bioavailable nitrogen from atmospheric N_2 gas. Here we explore weathering of ammonium contained in felsic igneous rocks as an alternative source. We find that this process could have supplied 10^8–10^9 mol yr^{-1} of bioavailable N to surface environments in the early Archean, leading to dissolved concentrations of 0.023 ± 0.017 μM in freshwater and 0.01–0.1 μM in seawater. In terrestrial settings, evaporation paired with elevated N supplies from locally enriched felsic bedrock may have led to concentrations approaching 1 μM. Rock weathering would thus have constituted a smaller flux than the sum of all proposed high energy sources of fixed N, but with the major benefit that it was reliably present, especially in terrestrial settings. Weathering of differentiated igneous rocks should thus be considered in models of the emergence of life on Earth and beyond.

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Introduction

All life as we know it requires reduced nitrogen (N) in ammonic form to build amino acids, DNA, RNA and other fundamental biomolecules that fuel biochemical processes in living cells. Therefore, the requirement for ammonic N most likely extends back to the prebiotic world. Today, the major source of ammonium to the biosphere is biological N_2 fixation with a flux of *ca.* 1.1×10^{13} mol yr^{-1} (Wang *et al.*, 2019); however, this metabolism would initially not have existed. Hence prebiotic reaction networks and perhaps the earliest life forms in the Eo- to Palaeoarchean (4.0–3.2 Ga) would have relied on abiotic mechanisms to supply ammonic N.

A number of high energy planetary-scale processes have been identified that can convert atmospheric N_2 gas — the largest reservoir of N at Earth's surface — into bioavailable forms. These include lightning (10^9–10^{10} mol yr^{-1} in the Archean) (Kasting and Walker, 1981; Navarro-González *et al.*, 1998), volcanic eruptions (10^9–10^{11} mol yr^{-1}) (Mather *et al.*, 2004), impact shock waves (10^{10}–10^{11} mol yr^{-1}) (Kasting, 1990; Nakazawa *et al.*, 2005), photochemical reactions (10^9–10^{11} mol yr^{-1}) (Tian *et al.*, 2011; Wogan *et al.*, 2023) and hydrothermal vents (10^9–10^{12} mol yr^{-1}) (Brandes *et al.*, 1998; Smirnov *et al.*, 2008). Of these, lightning, volcanism, and impact shocks generate N oxides, which require conversion into ammonium by redox reactions involving ferrous iron (Wang *et al.*, 2023). While all of these are plausible N sources that almost certainly contributed to the early N cycle, they have in common that they are spatially and/or temporally restricted, making them unreliable for a nascent biosphere that attempts to expand into new ecological niches. To overcome this limitation, we quantitatively explored the hypothesis that rock weathering contributed a continuous supply of bioavailable ammonium on the early Archean Earth.

On the modern Earth, rock weathering has been found to generate a N flux of *ca.* 0.8–1.3×10^{12} mol yr^{-1} from land to sea (Houlton *et al.*, 2018), equivalent to up to 10 % of the biological N_2 fixation flux. However, this value is not directly transferrable to the early Earth for several reasons: (1) the N content of modern bulk continental crust is elevated due to storage of biomass in (meta-)sedimentary rocks and granitoids (Mikhail *et al.*, 2024), (2) the mass of exposed continental crust may have been smaller in the past (Dhuime *et al.*, 2015), and (3) continental crust probably had a different chemical composition with a higher degree of maficity than it does today (Greber *et al.*, 2017). Here, we account for these three parameters to derive plausible upper and lower bounds for a N weathering flux and resulting fluid ammonium concentrations in the early Archean, enabling us to determine if this flux could have contributed to the origin and sustenance of first life.

The Early Archean Crustal N Reservoir

Similar to other cations, N as ammonium (NH_4^+) contained in crustal rocks can be liberated by weathering as demonstrated from modern soils (Dahlgren, 1994). Quantifying the global prebiotic N weathering flux requires knowledge of the amount of lattice-bound N in Earth's first land masses. Today's continental crust contains *ca.* $1.7 \pm 0.1 \times 10^{18}$ kg of N (nearly half as much as the atmosphere) with an average concentration of 150 ± 12 μg g^{-1}

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in the upper crust (Johnson and Goldblatt, 2015). Much of this rock-bound N is hosted in (meta-)sedimentary rocks and sediment derived granitoids. In contrast, on the early Earth, prior to the origin of life, N in crustal rocks would have been derived from purely magmatic sources, *i.e.* not involving biomass. The earliest felsic rocks in the continental crust were formed by (1) re-melting of mafic igneous rocks. or (2) magmatic differentiation of mafic melts. Both processes result in incompatible element-enriched melts (*e.g.*, elevated concentrations of large ion lithophile elements). During magmatic differentiation, N in the form of ammonium (NH_4^+) behaves similar to LILEs, such as potassium (K^+), due to their similarity in charge and ionic radius. It has been shown that magmatic differentiation can lead to a significant enrichment of igneous sourced (primary) N in continental crust-building felsic igneous rocks (Boocock *et al.*, 2023a). For example, Hekla volcano on Iceland shows an increase of N from 3.3 μg g^{-1} in basalt to up to 23 μg g^{-1} N in rhyolites (Fig. 1a). The concentration in the felsic end member of Hekla implies that between 8.06×10^{16} kg and 1.36×10^{17} kg of N in the continental crust today could be derived from magmatic differentiation of mantle melts. By comparison with the total mass of N in the granitic (filtered to >60 % SiO_2) upper continental crust (2.62×10^{17} kg N) (Johnson and Goldblatt, 2015), igneous N can account for between 31 to 52 % of felsic stored N. In igneous rocks, N is most likely hosted in the form of lattice bound ammonium in feldspars and micas (Boocock *et al.*, 2023b; Honma and Itihara, 1981), and both phases break down during weathering and release cations into solution (Wilson, 2004).

Collectively, these observations show that the igneous crustal N reservoir is important to consider as a nutrient source to prebiotic processes and early biotic communities. Direct constraints on the N concentration in early Archean crustal rocks are so far lacking, but we can derive plausible lower and upper bounds. First, Earth's mantle is estimated to contain *ca.* 0.84 ± 0.43 μg g^{-1} N (Marty, 2012) and modern oceanic crust contains 1.4 ± 1.3 μg g^{-1} (Johnson and Goldblatt, 2015). Given the magmatic differentiation trends documented from the modern Hekla suite (Fig. 1a), the N concentration of early Archean felsic rocks was likely higher than in these two reservoirs. We will therefore proceed with 2 μg g^{-1} as a lower bound for the N concentration of the first continental crust. Second, Neoarchean peraluminous granites have been found to contain 8 ± 5 μg g^{-1} N (Mikhail *et al.*, 2024). These granites must have involved some degree of sediment melting during their formation to develop the peraluminous character. As sediments are likely to contain biomass and hence biogenic N, the mean concentration of 8 μg g^{-1} in these peraluminous granites is thus a plausible upper limit for N in the prebiotic continental crust. Importantly, average crust is not purely felsic but intermediate in composition, *i.e.* involving a combination of mafic and felsic rocks. Taken together, we therefore proceed with a mean of 5 ± 3 μg g^{-1}, but we also consider enrichments up to 23 μg g^{-1} later in the discussion to account for magmatic processes seen at Hekla today (Boocock *et al.*, 2023a). We could alternatively use the estimated K content of early Archean crust (*ca.* 1.5 wt. % K_2O) (Greber *et al.*, 2017) and scale it by the N/K ratio of a purely igneous system such as Hekla volcano (*ca.* 8 μg g^{-1} *per* 1 wt. % K_2O; Fig. 1a) to derive an Archean crustal N concentration of about 12 μg g^{-1}, but this value would be slightly higher than the average of Neoarchean peraluminous granites. Hence, the approach described above is more conservative.

Prebiotic N Weathering

As noted earlier, N in igneous rocks is largely hosted in feldspars and micas, due to the similarity in charge and size between ammonium and K. Feldspars and micas are also the major hosts of K in the crust, which leads to a strong covariance between the two elements in both igneous and metamorphic suites (Boocock *et al.*, 2023a; Busigny and Bebout, 2013). Hence, the well established rate at which K is released into the environment during weathering can be used to derive the rock weathering flux of ammonium (*i.e.* independent from biomass weathering). The modern upper crust contains on average 2.41 ± 0.22 wt. % K (Greber *et al.*, 2017), and modern rivers carry 1.58 ± 0.34 μg g^{-1} K in solution (Wang *et al.*, 2021). With a global river water flux of $3.74 \pm 0.78 \times 10^{16}$ L yr^{-1} (Collins *et al.*, 2024), this leads to a modern global riverine K flux of $5.91 \pm 1.77 \times 10^{13}$ g yr^{-1}. For a modern continental N concentration of 150 ± 12 μg g^{-1} (Johnson and Goldblatt, 2015), this would imply a N rock weathering flux of $3.7 \pm 1.2 \times 10^{11}$ g yr^{-1}, or $2.6 \pm 0.8 \times 10^{10}$ mol yr^{-1}. However, it is important to note that in the modern crust, some fraction of N is bound to organic matter in shales (Johnson and Goldblatt, 2015), meaning that it is not hosted in potassic silicate minerals and may weather at a different rate than K. Therefore, scaling N rock weathering by K weathering is inaccurate for the

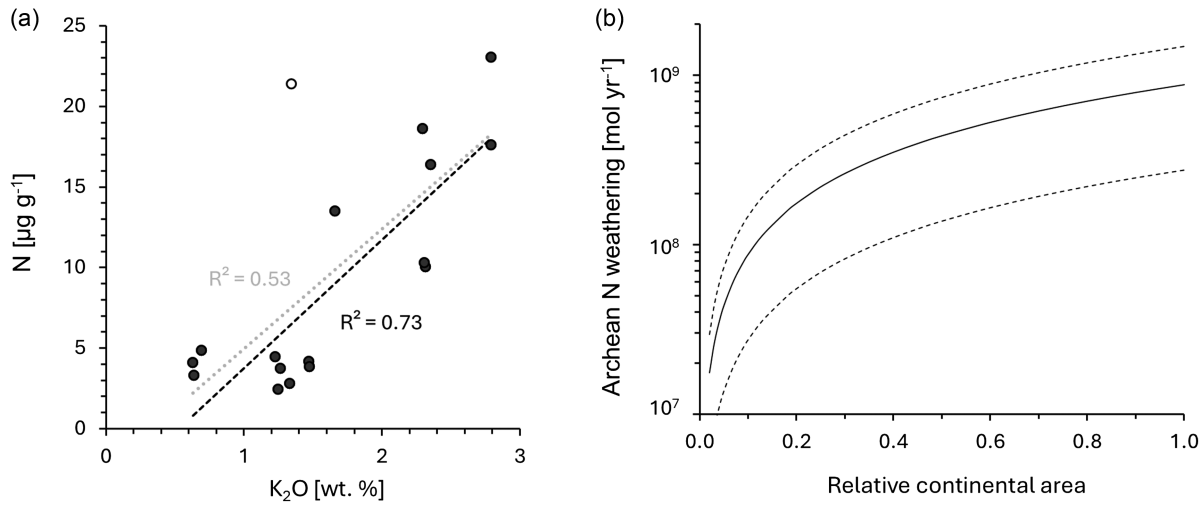

Figure 1 **(a)** Whole-rock data showing the N *versus* K concentrations in aphyric lavas from Hekla volcano (Boocock *et al.*, 2023a). The correlation coefficient of all data ($R^2 = 0.53$, grey trendline) increases ($R^2 = 0.73$, black trendline) if one outlier is excluded (unfilled point). **(b)** Calculated N weathering flux for the early Archean (see text for derivation).

modern crust. In contrast, on a prebiotic world, a biogenically derived organic reservoir would have been absent, such that K weathering, which tracks only silicate bound N, can provide a useful benchmark. We therefore proceed with our K-based approach, scaled by the average size of the early Archean continental N reservoir of 5 ± 3 μg g^{-1} derived above.

Implicit to our approach is the assumption that N-bearing silicates weather at the same rate as K-bearing silicates over geological time scales, including formation and dissolution of secondary minerals, such that we can calculate the N weathering flux as $F_{\text{N-weathering}} = [\text{N}]_{\text{crust}}/[\text{K}]_{\text{modern-crust}} \times F_{\text{K-modern}}$, where $[\text{N}]_{\text{crust}}$ is the assumed concentration of N in the ancient crust, $[\text{K}]_{\text{modern-crust}}$ is the concentration of K in the modern crust, and $F_{\text{K-modern}}$ is the modern weathering flux of K. We are also assuming that weathering rates overall have been the same on the early Earth as today, but we acknowledge that this is uncertain as global climate may have differed in ways that are poorly constrained.

Results and Discussion

The approach outlined above yields a global average N weathering flux of $8.8 \pm 6.0 \times 10^8$ mol yr^{-1} ($1.2 \pm 0.8 \times 10^{10}$ g yr^{-1}). We emphasise that potential sinks of ammonium are not considered and may reduce this value. These include volatilisation of ammonia (NH_3), potential photochemical destruction and other reactions, all of which are difficult to quantify. However, at least 21 % of precipitation recharges groundwaters today (Xie *et al.*, 2024), and ammonium contained in this groundwater reservoir would have experienced only limited ammonium loss by volatilisation and photooxidation. Also, in surface waters, UV radiation would have been at least partly attenuated. We therefore proceed with the flux calculated above, while highlighting that additional work is needed to quantify ammonium loss processes from weathering environments on an abiotic world.

Using these constraints, the concentration of dissolved ammonium in river waters would be 0.023 ± 0.017 μM, compared to 0.7 ± 0.3 μM with our modern weathering flux of $2.6 \pm 0.8 \times 10^{10}$ mol yr^{-1} (see above). We can further scale the calculated early Archean flux by a smaller continental landmass, assuming that the river water flux scales linearly with land area. For example, for 10–20 % of modern land exposure results in a global average ammonium flux estimate of $0.9–1.8 \times 10^8$ mol yr^{-1} (Fig. 1b). Also, the riverine water concentration may vary if rainfall is not uniformly distributed, or if the rain rate and hence river water flux were significantly different in the past, depending on global temperature. With the modern river water flux of $3.74 \pm 0.78 \times 10^{16}$ L yr^{-1} and a global land surface area of 1.48×10^{14} m^2, the global average rain rate is 253 L m^{-2} yr^{-1}. The lower the rainfall, the higher the resulting dissolved ammonium concentration, as ions are less diluted (Fig. 2a), consistent with observations in modern rivers (Hung *et al.*, 2020). At very high rain rates weathering may become erosion limited. This threshold is dependent on climate and tectonics and unknown for the early Earth. However, at low rain rates where N concentrations would be highest this concern would not be relevant.

Lastly, we can use the global weathering flux to calculate the resulting marine ammonium reservoir, using a simple box model of an abiotic N cycle from a previous study (Stüeken, 2016). Here, the source is the total abiotic ammonium input while sinks include ammonium adsorption to clays and pH dependent ammonia volatilisation with an assumed ocean turnover rate of 1000 yr. The results show that for a total source flux of 10^8–10^9 mol yr^{-1} derived from rock weathering the steady state concentration of ammonium in the ocean would have been 0.01–0.1 μM (Fig. 2b). Additional ammonium may have been contributed by weathering of oceanic crust not explicitly considered in this study; this flux was likely smaller than the continental flux, given the low N content of mafic rocks (see above). By contrast, using the proposed source fluxes from lighting, volcanism, impact shocks, photochemistry and hydrothermal vents (10^9–10^{12} mol yr^{-1}) and assuming complete conversion to ammonium in all cases would yield dissolved marine ammonium concentrations of *ca.* 1–1000 μM in the global ocean.

Our calculated rock weathering flux of ammonium is smaller than proposed fluxes based on high energy processes in the atmosphere or in the deep ocean (Table 1). Nevertheless, this flux may have played a role in the origin and early evolution of the biosphere because rock weathering provides (a) a constant supply of fixed N, unlike sporadic high energy events, (b) N in the form of ammonium, circumventing

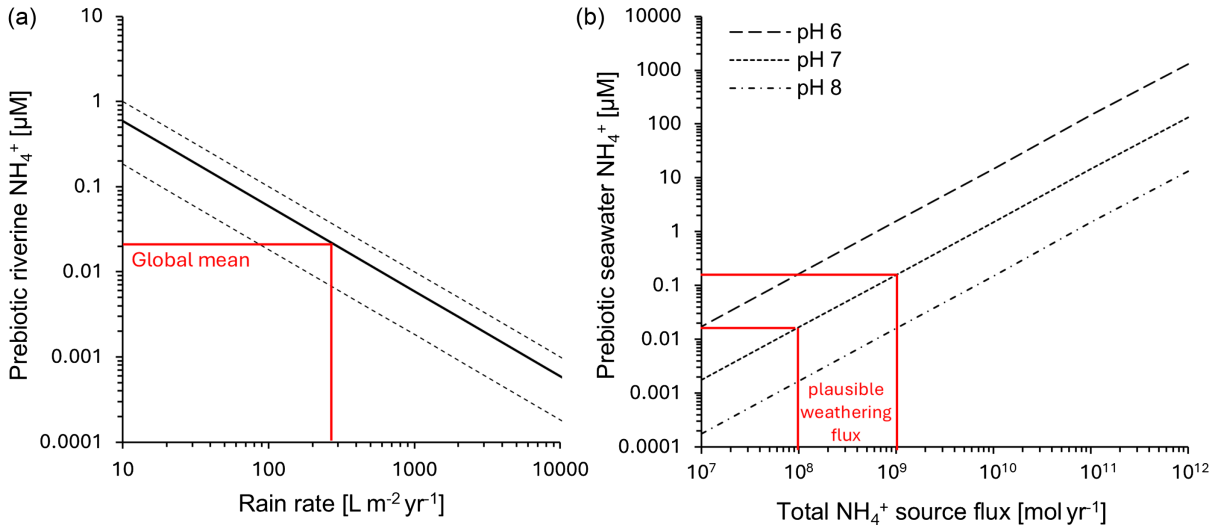

Figure 2 Fluid concentrations on the prebiotic Earth for **(a)** average river waters and **(b)** average seawater. The latter includes sinks due to clay adsorption and ammonia degassing (Stüeken, 2016). The pH range from 6 to 8 includes estimates of early Archean seawater pH of *ca.* 6.5 (Halevy and Bachan, 2017).

Table 1 Calculated prebiotic fluxes of fixed N in mol yr^{-1}.

Flux type	Magnitude (mol yr^{-1})	Dominant product	Supply rate	Ref.
Lightning	10^9–10^{10}	Nitrogen oxide(s)	Sporadic	1
Volcanic eruptions	10^9–10^{11}	Nitrogen oxide(s)	Sporadic	2
Impact shock waves	10^{10}–10^{11}	Nitrogen oxide(s)	Sporadic	3
Photochemistry	10^9–10^{11}	Hydrogen cyanide	Constant	4
Hydrothermal vents	10^9–10^{12}	Ammonium	Constant	5
Crustal weathering	10^8–10^9	Ammonium	Constant	6

1 = Navarro-González *et al.* (1998), Kasting and Walker (1981); 2 = Mather *et al.* (2004); 3 = Kasting (1990), Nakazawa *et al.* (2005); 4 = Tian *et al.* (2011), Wogan *et al.* (2023); 5 = Brandes *et al.* (1998), Smirnov *et al.* (2008); 6 = this study.

the need of reacting atmospheric N oxides with ferrous iron, and (c) fixed N available on land, in lacustrine settings that were cut off from deep marine hydrothermal ammonium sources. Furthermore, the riverine ammonium flux could have become concentrated in local evaporitic ponds. It may also have been further elevated in the vicinity of N-enriched felsic rocks, such as at Hekla volcano or felsic plutons, where evolved silicic rocks can show N concentrations that are *ca.* 5 times higher than the global crustal average assumed in our model for the early Archean (Boocock *et al.*, 2023a). For example, evaporation by a factor of 10, paired with locally enriched rocks by a factor of 5, could yield dissolved ammonium concentrations approaching 1 μM in lakes. In comparison, the modern deep ocean contains 30 μM of dissolved nitrate, with much lower concentrations down to a few μM or less in surface waters (Gruber, 2008). Fixed N concentrations around 1 μM are thus significant for biological processes. Lastly, we emphasise that rock weathering would operate on any Earth-like planet with a differentiated crust and a hydrosphere. The ability of this process to generate significant levels of dissolved ammonium thus speaks to the origin of habitable biochemical conditions on other worlds.

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