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# *Interactive comment on* "A new synthesis for terrestrial nitrogen inputs" *by* B. Z. Houlton and S. L. Morford

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Interactive comment on "A new synthesis for terrestrial nitrogen inputs" by B. Z. Houlton and S. L. Morford W.

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Review of Houlton and Morfort A new synthesis for terrestrial nitrogen inputs Soil 2014-29

Author response in bold.

The authors provide a nice summary of the sources of reactive nitrogen on the Earth's

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land surface, covering the well-known inputs by N-fixation and industrial fertilizer, and focusing on the nitrogen derived from the lesser-known pathway of rock weathering. The latter was recognized by these workers and their colleague, Randy Dahlgren, at UC Davis only within the past couple of decades.

The authors make a strong case for the importance of rock-derived nitrogen in a variety of regional settings. Surprisingly, they do not mention that in the preindustrial world, rock-derived N (20 TgN/yr) may have been in the same order of magnitude globally as nitrogen from biological fixation (60 TgN/yr; Vitousek et al. 2013), a major modification of a paradigm of biogeochemistry. Now, of course, human production of nitrogen fertilizer (∼140 TgN/yr) and cultivation of leguminous crops (60 TgN/yr) dwarfs both natural sources.

Author: We agree that the role of rock N inputs to the global N supply, both under modern and pre-industrial input scenarios, is an area that deserves more research. Not only are rock N inputs potentially of similar magnitude to N fixation, but because the factors that regulate these two N input processes at biome and landscape scales differ, our new synthesis suggests where rock N inputs may dominate in a given ecosystem site.

Of course, human actions have more than doubled the amount of fixed N inputs to the land system. However, the importance of rock N to the remainder of the terrestrial biosphere, where anthropogenic N inputs are more limited, is still uncertain, though likely meaningful (at least ∼20 Tg N/yr).

We have added several sentences in the introduction to address the potential global N input magnitudes:

“Geochemical models have pointed to the importance of N weathering in regulating atmospheric N2 over deep time (Berner, 2006). The burial of fixed N in marine environments (∼25 Tg yr-1 , Gruber and Galloway, 2008 ) cannot be compensated by solely volcanic degassing (∼ 0.4 Tg yr-1, Busigny et al. 2011), suggesting that the

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majority of the N transferred to the crust must be recycled via rock uplift and weathering. This implies that global rock N inputs may be of similar magnitude to lower-bound estimates of biological N fixation in natural terrestrial sites (58 Tg yr-1, Vitousek et al. 2013)."

This is an interesting and readable paper that I could easily imagine as an assignment in a graduate class in biogeochemistry. Aside from atrocious misuse of the hyphen, the authors only give me a few points to quibble:

Author: We have substantially edited the text to further improve the clarity of the manuscript. This includes removing many of the em-dashes. Thank you for the advice.

Line 159. With such a robust literature documenting ecotypic differentiation of enzyme temperature optima in plants and soil microbes, I am surprised that the same is not true of nitrogenase. Or at least, that no comment was made on this.

Author: We have added the following text: "There is little evidence for acclimation of N fixation across tundra to tropical climates, perhaps owing to the complex nature of the nitrogenase enzyme (Houlton et al. 2008). Rather, the integrated data in Fig. 1 can be fitted to a single Arrhenius function with slope that falls between the steep temperature-dependence of the nitrogenase enzyme and the less pronounced temperature sensitivity of photosynthesis."

Line 214. Free-living (asymbiotic) N-fixation receives scant treatment in this paper, even though at rates of 1 to 5 kg/ha/yr, it may account for as much as 1/3 of the biological nitrogen-fixation on land. In particular, it would be nice to comment on the importance of free-living nitrogen fixation, especially in certain desert environments and their soil crusts.

Author: We have addressed free-living rates globally and in desert crusts.

We have addressed this point in the following text:

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“This is pronounced in desert ecosystems where patch-scale heterogeneity in soil-crust communities and seasonality in moisture and temperature alter spatial patterns of N fixation and nutrient cycling (Belnap, 2002).”

“Studies using foliar 15N/14N in arid sites have suggested similarly high rates of N fixation (9 to 22 kg N/ha/yr) in Prosopis glandulosa (mesquite) stands (Geesing et al., 2000).”

“Free-living rates of fixation in rocks and soil are lower than symbiotic ones, but the widespread distribution of cryptograms, and the capacity of such organisms to respond rapidly to change, means that this functional group is globally important, perhaps accounting for up to 50% of natural terrestrial N fixation (Elbert et al., 2012).”

Lines 272-273. The increase in rock weathering expected with plant growth at elevated concentrations of atmospheric CO2 may be slight, inasmuch as plants and soil microbes normally maintain pCO2 at high levels in the soil pore space, so the increment with rising atmospheric CO2 is likely to be small, although nevertheless significant over geologic time (Andrews and Schlesinger 2001).

Author: We agree that the direct changes in soil pCO2 will likely be small under elevated CO2, but the integrated effect of higher belowground C allocation contributes to enhanced root production, reactive mineral surfaces, and acidity that can significantly increase weathering rates at both short and long timescales. We’ve altered the text reflect this combined effect on mineral weathering.

Line 341. Presumably the authors mean “catenae” Line 354.

Author: Fixed.

Line 354. The huge pool of nitrogen in the Earth’s crust is not so easily interpreted to indicate that N-fixation has exceeded denitrification through geologic time. Some of this nitrogen may have never circulated in the biogeochemical cycles at the Earth’s surface. Rather, it would be interesting to estimate the amount of nitrogen being sub-

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ducted in sedimentary rocks passing into the Earth's mantle versus the amount that is being degassed as juvenile nitrogen by volcanoes. Recent estimates suggest that volcanic emissions (78 to 123 x 109 gN/yr) are less than subduction (330 to 960 x 109 gN/yr), suggesting a net entrainment of N in the Earth's mantle. See Schlesinger and Bernhardt (2013, p. 456) for references.

Author: We acknowledge that there may be some confusion here because Mantle fixed-N reservoirs may be substantial, and do not contribute substantially to surface (biogeochemical) N cycling. However, excluding Mantle reservoirs, the 99% statement still holds.

Further, 75% of the continental crust N reservoir is found in sedimentary and meta-sedimentary rocks, and most of this N derived from burial of organic matter in Phanerozoic rocks (See Bebout et al, 2014). The N content of the early continental crust was likely on the the order of 1016 kg (assuming the N content of the upper mantle is good analogy for early continental crust; see Marty 1995 - Nature), while the modern N reservoir in continental crust is 1018 kg, an increase of two orders of magnitude. While a fraction of crustal N may have originated prior to the rise of biological N fixation (See Goldblatt et al. 2009 – Nature Geoscience), the majority of this N has accumulated as a result of organic matter burial in sedimentary rocks derived from terrestrial and oceanic N fixation.

With respect to N exchange with the mantle, the net transfer of N from earth surface + crust reservoirs to the mantle is estimated at 0.9 Tg yr-1 (see Busigny et al, 2011), and has been hypothesized (controversially) to be substantially larger during early earth evolution (Goldblatt et al, 2009). However, with respect to a planetary N cycle, the primary imbalance appears arise from the 15 – 35 Tg N that is buried in marine environments annually. Given that combined volcanic emissions are ∼0.5 Tg yr-1, more than 90% of the N flux into the crust reservoir must be recycled to the biosphere to maintain atmospheric N reservoirs over geologic time.

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New text reads:

“Rocks contain ∼99% of Earth’s fixed N (Schlesinger, 1997), even when excluding mantle reservoirs that interact sparingly with earth surface processes (Bebout et al., 2013). Approximately 75% of the fixed N reservoir within the continental crust is found in sedimentary and meta-sedimentary rocks (Goldblatt et al., 2009), primarily reflects higher rates of N fixation compared to denitrification to N2 gas over Earth history. Nitrogen concentrations are much higher in sedimentary/meta-sedimentary than igneous parent materials, though either class can contain appreciable geological N (Holloway and Dahlgren, 2002). Further, reservoirs of geological N can occur as silicate-bound NH4+, organic-N in sedimentary organic matter, or nitrate in evaporites. Variation in both the amount and form of rock N is controlled by local depositional environments, the degree of biological and thermal digenesis in sedimentary basis, and the degree of N volatilization during metamorphism (Bebout and Fogel, 1992; Hedges and Keil, 1995; Hedges et al., 1999; Boudou et al., 2008). Rock-bound nitrate can be seen in desert/arid ecosystems where hydrological losses are minimal and nitrate accumulates at depth or in the surface of caliche deposits (Walvoord et al., 2003). On average, Holloway and Dahlgren (2002) found that the parent material factor is a strong driver of rock N contents, with trace amounts of N found in cratonic assemblages to >20,000 ppm N in sedimentary rocks such as coal. Generally, N enrichment is highest among fine-grained siliciclastic rocks (i.e. shales, mudstones), and their low-grade metamorphic counterparts (i.e. slate, phyllite, and mica-schist). These rocks comprise ∼30% of earth’s continental surfaces (Durr et al., 2005) and have an average N concentration equal to 700 – 1000 mg N kg-1 (Goldblatt et al., 2009; Morford, 2014).”

Line 419. Schlesinger et al. (1998) document phosphorus deficiencies in recent volcanic soils on Krakatau. These do not appear to be related to low P concentrations as much as to a stoichiometric deficiency of P relative to high concentration of N that was accumulated in these soils by cyanobacteria, which are reported to have colonized immediately after the 1883 eruption.

Author: Thanks, this is an interesting paper that escaped our radar. The high rates of N accumulation in Krakatau seem to be consistent with free-living fixation, although Schlesinger et al.'s 1998 analysis is inferential rather than direct. We have modified the text as follows:

"In both newly formed volcanic (Vitousek, 2004) and de-glaciated sediments (Chapin et al., 1994) cyanolichens are among the earliest colonizers, with direct and indirect evidence for significant free-living N fixation rates in fresh parent material (Schlesinger et al., 1998; Crews et al., 2001)"

Line 486. These air-borne agricultural losses of N represent an input to ecosystems that are downwind, but globally they merely represent a recycling of N added to the land surface by the application of industrial fertilizer

Author: Agreed. New sentence added to text:

"From a global budget perspective, agricultural emissions of NOx and NH3 comprise a large-scale recycling term, despite representing a new N input to downwind ecosystems."

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Interactive comment on SOIL Discuss., 1, 497, 2014.