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Journal

Biogeochemistry., 132(1-2)

ISSN

0168-2563

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Publication Date

2017

DOI

10.1007/s10533-016-0286-5

Peer reviewed

Erosional redistribution of topsoil controls soil nitrogen dynamics

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Received: 9 May 2016 / Accepted: 28 November 2016 /
Published online: 26 December 2016

Abstract In recent years, the role of soil erosion on terrestrial carbon sequestration had been the focus of a growing number of studies. However, relatively little attention has been paid so far to the role of erosion on the lateral distribution of soil nitrogen (N) and the role of geomorphic processes on soil N dynamics. Here, we present primary data on the stock of nitrogen in soil and its rate of erosion at a relatively undisturbed, zero-order watershed in northern California. Erosion transports $0.26\text{--}0.47\text{ g N m}^{-2}\text{ year}^{-1}$ from eroding slope positions (Summit and Slope), and about two-thirds of the eroded N enters depositional landform positions (Hollow and Plain). Our results show that depositional-position soil profiles contain up to 3 times more N than soil profiles in the eroding positions. More than 92% of all soil nitrogen was chemically bound to soil minerals in all the landform positions, compared to 2–4% each found in the free light and occluded light fractions. Nitrogen associated with the free light

light fractions and the mineral-associated dense fractions is likely to be protected from gaseous and dissolved losses. On average, we found that soil N has mean residence time of 694 years in eroding landform positions, compared to 2951 years in depositional landform positions. Our results also show that microbial processing of organic matter exerts strong control on overall soil N storage and N stabilized through sorptive interactions with soil minerals only in poorly drained depositional landform positions. Soil erosion exerts important control on stock, distribution, and long-term fate of soil N in dynamic landscapes.

Keywords Soil erosion · Soil nitrogen · Soil organic matter · Coupled C and N cycling

Introduction

Soil erosion and terrestrial sedimentation regulate stock and fluxes of essential elements in the soil system. Soil erosion laterally redistributes about 75 Gt of soil annually (Stallard 1998). Over the last two decades, a growing number of studies have been investigating the effects of erosion on SOM dynamics and biogeochemical cycling of essential elements, especially carbon (C) (for example: Berhe 2012;

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Berhe et al. 2007, 2008, 2012, 2014; Boix-Fayos et al. 2009; Dlugosz et al. 2012; Doetterl et al. 2012a, b, 2016; Dymond 2010; Harden et al. 1999, 2008; Hu et al. 2016; McCorkle et al. 2016; Sanderman and Chappell 2012; Stacy et al. 2015; Van Oost et al. 2007, 2009, 2010, 2012a, b; VandenBygaart et al. 2012; Wang et al. 2013; Zhang et al. 2012). In comparison, less attention has been paid to how erosion affects the dynamics of soil nitrogen (N) (Gimeno García et al. 2000; Hilton et al. 2013; Kaye et al. 2003; Palis et al. 1997; Parfitt et al. 2013; Quinton et al. 2010; Stacy et al. 2015; Stewart et al. 2014). This is despite the fact that studies from as far back as 1936 have highlighted the role of topography and associated transport of topsoil by soil erosion on the biogeochemical cycling of N (Aandahl 1948; Houlton and Morford 2015; Jenny 1941b; Jenny et al. 1949; Milne 1936; Schimel et al. 1985).

Important differences in soil N dynamics are to be expected even between eroding and depositional landform positions the same catchment. In upland,

Responsible Editor: Steven Perakis.

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fraction in topsoil is particularly susceptible to loss by soil erosion. By comparison, soil N associated with the aggregate-protected occluded

eroding landform positions, erosion affects soil N dynamics through its effects on loss of N via direct removal of soil mass and light-density carbonaceous materials such as particulate OM from the soil surface (Gregorich et al. 1998; Lal 2003; Rumpel et al. 2006b) and dissolved losses of N with runoff (Schuman et al. 1973). On the other hand, on lower-lying depositional landform positions, erosion is likely to influence soil N dynamics through delivery of N-rich topsoil from upslope positions, its effects on net primary productivity, burial of topsoil, associated changes in rates of decomposition/stabilization of soil organic matter, and N mineralization and leaching (McCorkle et al. 2016; Stacy et al. 2015).

Nitrogen is an essential plant nutrient that imposes strong limits on primary production and biogeochemical cycling of elements in most terrestrial ecosystems (van Breemen 2002; van Breemen et al. 2002). There is a longstanding recognition that availability and flow of water controls multiple processes in the N cycle (Cirimo and McDonnell 1997; Palis et al. 1997). It is also recognized that the N cycle is characterized by a strong transport component locally within soil profiles (Hart et al. 1993), and most of the land-fixed N is laterally transported and denitrified in aquatic environments (Gruber and Galloway 2008; Meybeck 1982). However, most studies on lateral transport of N in terrestrial ecosystems focus on fluxes of dissolved spaceorganic or inorganic forms of N from the soil system to streams, rivers, and estuaries (Meybeck 1982; Schlesinger et al. 1999). Relatively little attention has been paid to the transport of particulate N (Hilton et al. 2013; Likens and Bormann 1974). It is estimated that the export of eroded particulate N is roughly equivalent to the rate of total dissolved N loss at the catchment scale (Lewis et al. 1999). There is currently very little data on spatial distribution of soil N, the role of erosion in the lateral distribution of particulate N in hillslope environments, and the implications of particulate N erosion for organic matter dynamics even though previous studies had suggested that up to 92% of N lost from eroding watersheds could be with sediment in runoff (Schuman et al. 1973).

Globally, about 70–90% of eroded soil material never makes it to the ocean: it is deposited in different terrestrial depositional settings (Stallard 1998) that are characterized by variable mechanical, hydrological, and microclimatological conditions

that affect soil C and N dynamics (Berhe 2013). Because cycling of N in the terrestrial ecosystem is sensitive to changes in microclimate (Brady and Weil 2008), the fraction of eroded N that is deposited in different terrestrial depositional landform positions can be rendered unstable during or after erosional transport. This is likely because soil N undergoes continuous transformation and loss in/from soil through the combined processes of nitrification and denitrification, leaching, (Matheny and Hunt 1983), and runoff. Increased rates of organic N mineralization during detachment and transport phases of soil erosion are likely to render N in particulate and dissolved forms susceptible to loss (Quinton et al. 2010). It is also likely that gaseous forms of N can be released from soil and sediments by photooxidation (Mayer et al. 2012) during or after transport. On the other hand, stabilization of N in soil occurs through physical and chemical association of organic N containing compounds with soil minerals, and ammonium fixation in soil, which along with burial protect N from loss (Quinton et al. 2010).

Quinton et al. (2010), estimated that global soil N erosion rates from agricultural systems were 23–42 Tg N year⁻¹. In comparison, 112 Tg of N is annually applied to agricultural lands as chemical fertilizers; 75 Tg year⁻¹ of N is removed in harvested crops; and 23–30 Tg N year⁻¹ is exported from watersheds to river systems as particulate N. Most N in soil is derived from biological fixation (Matheny

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and Hunt 1983), where it rapidly accumulates and is retained in stable pools of soil organic matter, in forms that are not readily accessible by the soil microbial community (Kaye et al. 2003). Determining the role of erosion in soil N is important, because erosion is the only mechanism by which stable, mineral-associated organic N can be redistributed in large quantities (Berhe et al. 2012; Berhe and Kleber 2013; Kaye et al. 2003). The cycles of C and N (Thornton et al. 2009), as well as microbial and transport processes (Rockhold et al. 2004) in the terrestrial ecosystem, are tightly coupled to determine the persistence of both C and N in dynamic ecosystems experiencing lateral distribution of topsoil material by erosion.

The rate of lateral distribution of N and loss of N in dynamic hillslope landscape is related to geomorphic, climatic, or ecological variables such as curvature, rainfall amount and timing, and rate of plant produc-

tivity (Houlton and Morford 2015; Peter John and Schlesinger 1990). In addition, studies have shown that geomorphology of the landscape exerts first order control on soil erosion (Houlton and Morford 2015; Roering et al. 1999), and the role of curvature on N cycling in dynamic landscapes has recently been confirmed in two studies that used $d^{15}N$ to trace dynamics of N in soils/sediments of eroding landscapes (Hilton et al. 2013; Weintraub et al. 2015). Furthermore, the rate of detachment and distribution of N with water erosion was found to depend on the extent and patchiness of plant cover (Turnbull et al. 2011). Climate—especially large seasonal variations in temperature and precipitation—was also related to the rate of N erosion and enrichment of N in eroded sediments (Ritchie et al. 2005). The rate of loss versus retention of eroded N from a given watershed also depends on spatial organization of the landscape, including the relative areas covered by eroding versus depositional landform positions and the prevailing hydrological and pedological properties in the soil profiles that dictate microbial biomass and activity, rates of biological N fixation, and rates of OM decomposition (Berhe 2012, 2013; Berhe et al. 2007, 2014; Berhe and Kleber 2013).

However, as of yet, despite these very important implications we still don't have accurate estimates of how much N is transported via erosion naturally, and its distribution in soils of dynamic landscapes and how soil N dynamics is related to overall SOM composition. The major objectives of this study are to: (1)

spacedetermine the N stock and rates of N erosion in an eroding watershed, (2) identify factors controlling the storage of different forms of N in eroding versus depositional landform positions, and (3) determine the relationship of N to changes in SOM composition in eroding versus depositional positions.

Methods

Study site

Samples for this study were collected from an undisturbed zero-order watershed (Heimsath et al. 1999) in the Golden Gate National Recreation Area (GGNRA) at Tennessee Valley (TV) in Marin County, California. The main source of disturbance in the site, grazing, was eliminated and the area has

been protected under the GGNRA at least since 1972 (Dietrich et al. 1995; Yoo et al. 2005a). A detailed description of the geomorphology, soils, and vegetation of the study site was previously provided in Berhe et al. (2008). Briefly, the site is characterized by Mediterranean climate region, with mean annual temperature of 14 °C and mean annual precipitation of 1200 mm. The study site is dominantly covered by Mediterranean annual grasses and a coastal shrub (*Baccharis pilularis*) that recently encroached into the area.

The physical and chemical properties of soils in TV vary along the toposequence where soils in the eroding slopes in TV are classified as Lithic Haplustolls, while those on the footslope are classified as Lithic Ustorthents and those on the depositional profiles are classified as Oxyaquic Haplustolls. The major parent materials for soils in TV are chert, greenstone, and Franciscan sandstone (Yoo 2003). The rate of soil erosion was previously determined to be between 50 and 130 g m⁻² year⁻¹, and the corresponding SOC erosion is about 2 g m⁻² year⁻¹ (Heimsath et al. 1999; Yoo et al. 2005c). Soil erosion in the study area is a result of gopher facilitated bioturbation and rain splash. In TV, bioturbation due to the activity of pocket gophers (*Thomomys bottae*) leads to extensive mixing of at least the top 30–40 cm of soil. Soil burrowed by gophers is transported downhill largely by diffusive mass transport (Berhe 2013).

The results presented in this work are from soil samples collected from four geomorphically distinct landform positions: two eroding (the Summit and the

spacebackslope, or Slope) and two depositional (a terrestrial depression or Hollow and an alluvial/colluvial Plain). We collected samples from six soil profiles at the Summit, eight at the Slope, six at the Hollow, and seven at the Plain. From each profile, soil samples

were collected at depth increments of 0–5, 5–15 cm, space($^{15}N/^{14}N$) isotopes in a sample and R_{std} is the ratio in a standard (Dawson and Brooks 2001).

The N inventory for the entire depth profile was calculated as:

$$N_{in} = \sum_{i=1}^n DZ_i \cdot \rho_i \cdot \delta 1 - R_{std} \cdot \delta \% N \cdot 100 \rho \cdot \delta 1 \rho$$

space then every 15 cm down to 120 cm, and finally every 30 cm, down to the soil-saprolite boundary. Soil

pH was measured in a 1:1 soil: water slurry, bulk density was determined by a combination of core and clod/ped methods (Blake and Hartge 1986), and particle-size distribution was determined using the hydrometer method (Sheldrick and Wang 1993). Cation exchange capacity (CEC) was determined using the barium- acetate calcium replacement technique (Janitzky 1986). Specific surface area (SSA) measurements were performed with N₂ gas adsorption at 77°K using a Micrometrics, Tristar 3000 automated gas adsorption analyzer (Micrometrics Instrument Corporation, Norcross, GA) and SSA calculated using the Brunauer–Emmett–Teller (BET) equation (Brunauer et al. 1938).

Carbon and nitrogen

For elemental and isotopic measurements of carbon (C) and nitrogen (N), the samples were air-dried, sieved to <2 mm, and ground using mortar and pestle to pass through a 100-mesh sieve. Percent C and N were measured with a Carlo Erba elemental analyzer, while the δ¹³C and δ¹⁵N were measured using a PDZ Europa Scientific 20/20 Mass Spectrometer and referenced to the Pee Dee Belemnite and atmospheric air standard. Similarly, harvested plant matter was oven-dried at 60 °C for 48 h and first ground with a Spex mill, and then with a Wig-L-bug mill until all fibrous material disappeared—before it was analyzed for C and N concentrations in a Carlo Erba elemental analyzer. The soil and vegetation δ¹⁵N and δ¹³C compositions were analyzed by continuous flow, using an elemental analyzer interfaced to an Iso-Prime100 mass spectrometer (detailed description of the sampling methodology of foliage is given in Berhe 2012). We express the stable isotopic compositions in ‰ notation (‰) as: δ¹³C or δ¹⁵N = $\frac{R_{sample}}{R_{std}} - R_{std} \times 1000$; where R_{sample} is the ratio of the heavy to the light C (C^{13}/C^{12}), N

space

where N_{inv} (g cm⁻²) is nitrogen inventory over the entire depth profile, DZ_i (cm) is thickness of i th soil layer, q_i is the bulk density (g cm⁻³), R_i is rock fraction, and %N is total nitrogen concentration in the

<2 mm fraction of soil in each layer.

The radiocarbon content of bulk SOM at all sampled soil layers was analyzed at the W. M. Keck

Carbon Cycle Accelerator Mass Spectrometry (AMS) Laboratory, following the methods of Trumbore et al. (1989) and Vogel (1992). The precision of the graphitization (by sealed tube Zn reduction) and AMS measurement on modern samples is about 2–3%, based on long-term duplicate analyses of standards and samples (Khosh et al. 2010; Xu et al. 2007). Carbon-14 content is expressed here as D¹⁴C (Stuiver and Polach 1977).

Erosional redistribution of soil N

The annual rate of N eroded from the Summit and Slope and the magnitude of N that is deposited in the depositional positions of Hollow and Plain was determined by dividing previously published rate of soil C erosion (Berhe et al. 2008) by the C:N ratio of the topmost soil layer in each landform position. Briefly, soil erosion rates were determined using cosmogenic isotopes by Heimsath et al. (1997). We used the rates of C erosion reported in the works of Yoo et al. (2005a, 2006; Yoo 2003). Yoo et al. (2005a, b, c, 2006) determined rates of C erosion for the study area by combining a hillslope sediment transport model with empirical soil C measurements and a topographic survey (using a total laser station) from which they derived slope gradient and curvature of the landscape across the zero-order watershed. In the works of Yoo et al. (2005a, b, c) soil transport in convex landform positions occurs as a result of diffusive mass transport by gophers and rain splash leading to a curvature-dependent rate of soil C erosion from eroding hillslope positions. On the other hand, rate of eroded C accumulation in the depositional Hollow, over 13,000 years of soil accumulation (since

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space last known major landslide event that excavated the Hollow's soil), was modeled based on changes in soil thickness due to deposition of soil eroded from the Summit and Slope positions. Yoo et al. (2005a, b, c) modeled the resultant soil C profile based on expected rates of plant productivity and decomposition. The rates of N erosion presented here are representative of the four landform positions of interest and do not capture the spatial variability of soil and nitrogen erosion/deposition rates in the all parts of the watershed (for spatial variability in rates of erosion, see Yoo (2003) and Yoo et al. (2005a, 2006).

Density fractionation

The bulk soil was separated into three density fractions—free light fraction (fLF), occluded light fraction (oLF), and dense fraction (DF)—using 1.7 g cm⁻³ sodium polytungstate [SPT, Na₆(H₂W₁₂O₄₀) solution according to the methods of Swanston et al. (2005), Strickland and Sollins (1987), and Golchin et al. (1994). Briefly, a 20 g soil sample was placed in a centrifuge bottle with 75 mL of SPT, and inverted gently (by hand) five times, let to settle undisturbed for 45 min, and centrifuged at 3500 rpm for 1 h. The free light fraction (fLF), which consists of large, undecomposed or partly decomposed plant fragments, was removed by aspirating floating material into a sidearm flask. To extract the oLF, the remaining soil mixture was mixed for 1 min with 75 mL SPT using a desktop mixer, sonicated for 3 min at 70% pulse, and centrifuged at 3500 rpm for 1 h according to the methods of Swanston et al. (2005). Preliminary tests that included sonication and wet sieving were used to determine that the applied ultrasonic energy level was sufficient to disperse all aggregates greater than 53 μ m in two representative samples. The extracted fLF and oLF were separately rinsed five times with deionized water, filtered to pass through a 0.8 μ m membrane filter, after which the filtrate was discarded and the fLF, oLF, and the residue (DF) were rinsed and oven-dried for 24–48 h.

Analytical precision, as determined by the standard deviation of repeated fractionation of the same sample (test carried out on 10 individual samples), was 2.04% for the recovery efficiency, 0.04% of total mass for the fLF, 0.11% of total mass for the oLF and 0.11% of total mass for the DF. In this procedure, it is not possible to determine the source of the fraction the N

spacelost during the fractionation steps. Hence, for the purpose of calculating the distribution of recovered N in the different fractions (fLF, oLF or DF), we assumed that the recovered mass was equivalent to total mass.

Data analysis

We express all data here using mean and standard error. Location means were compared using the Wilcoxon/Kruskall–Wallis nonparametric tests for means comparisons. We used a least squares nested ANOVA and compared pairs of means at the different depths and landform positions, using

the Tukey–Kramer HSD test to test for significant differences between means. Simple linear regression was used to assess the relationship between ¹⁵N in bulk soil and NMR derived SOM composition. For all statistical tests, a significance level of $p \leq 0.05$ was set before hand as the a level. All statistical tests were performed using JMP 6.0.0 software (SAS, Cary, NC).

Results

Table 1 summarizes the physical and chemical properties of soils in the eroding and depositional landform positions in TV. Soils in the eroding slope positions (Summit and Slope) are were less than 90 cm in thickness, compared to soils in the depositional landform positions (Hollow and Plain) that were as deep as 150 cm. The top 30 cm of all soils was very well mixed as a result of bioturbation by activity of pocket gophers, especially in the Summit, Slope and Hollow positions. Soils in the plain were most poorly drained, remaining waterlogged for most of the rainy season, compared to soils in the rest of the catchment that were well drained.

Soils in the depositional landform positions are characterized by finer texture, and higher bulk density, cation exchange capacity, and specific surface area. The pH of soils in the Hollow profile was slightly acidic (5.5–6.5 pH in water), whereas generally the soils in the other landform positions ranged from slightly acidic (5.4–6 pH in water) to neutral (6.5–7.2 pH in water) (Table 1).

A mix of annual grasses and Baccharis shrub covered all landform positions, except the Plain. The Plain, on the other hand, had denser vegetation cover space

Table 1 Soil properties at the eroding and depositional landform positions

Location	(m.a.s.l)	Sampled depth (cm)	Bulk density (g cm ⁻³)	pH in water	p C
Summit	105	0–5	1.3	5.9	5
		5–15	1.3	5.6	4
		15–30	1.5	5.6	4
		30–45	1.8	5.9	4
		45–69	1.8	6.6	5
Slope	80	0–5	1.6	5.4	4
backslope					

Hollow	50 footslope	5–15	1.6	5.4	concentration of N in biomass of annual grasses in the study area
		15–30	1.6	5.8	spacerranged from 0.72 to 0.95%, compared to 1.42–
		30–45	1.5	6.5	1.67% for Baccharis. The N:C ratio of vegetation was consistently higher for Baccharis than grasses in all
		45–60	1.8	6.9	landform positions, while there was no statistically significant difference in $\delta^{13}\text{C}$ of vegetation between
		60–75	1.8	6.7	grasses and Baccharis in all the landform positions.
		75–90	1.8	6.7	space
		0–5	1.5	5.5	space. Mean $\delta^{15}\text{N}$ was between 0.81 to 1.99% for grasses in
		5–15	1.5	5.5	the Summit, Slope, and Hollow positions compared to 3.79 % in the Plain, while the $\delta^{15}\text{N}$ of Baccharis was
		15–30	1.7	6.0	between 2.23 to 3.69% (Table 2).
		30–45	1.8	6.0	N storage in eroding versus depositional landform
Plain	45	45–60	1.6	6.2	positions
		60–75	1.8	6.0	The average N concentration (%N in the ~ 2 mm soil fraction) in the Plain and Summit was significantly higher than in the Slope and Hollow in the top 40
		75–90	1.9	6.0	cm of soil ($p \sim 0.05$). The %N in the Plain remained higher than the rest for the entire depth
		90–120	2	6.4	profile, while
		120–141	2.1	6.5	%N in the Summit showed a sharp drop below 40 cm.
		0–5	1.6	6.0	The total N inventory in the soil profiles in the depositional landform positions was up to 39 higher
		5–15	1.6	6.0	than in the eroding positions, largely because of the high N concentration in the top 40 cm and the deeper
		15–30	1.8	6.1	soils of the depositional landform positions compared to the eroding ones (Fig. 1).
		30–45	1.9	6.4	Erosional transport of soil N
		45–60	1.9	6.9	Erosion transports $0.25\text{--}0.47 \text{ g N m}^{-2} \text{ year}^{-1}$ from the eroding landform positions (Fig. 2). We found that the amount of N eroded from topsoil annually was equivalent to up to 10% of the N in above ground biomass in the eroding landform positions. If we average the rate of N erosion in the Summit and Slope, and average the rate of N deposition in the Hollow and Plain, we find that 57% of the N eroded from the
		60–75	1.9	6.9	spaceeroding positions is captured in the depositional
		75–90	1.8	6.9	positions (Fig. 2).
		90–105	2	7.2	Soil N:C ratios and $\delta^{15}\text{N}$ in eroding versus depositional positions
		105–120	2	7.2	The poorly drained soil profiles in the Plain had significantly higher (more positive) $\delta^{15}\text{N}$ and N:C ratio esp. in deep soil layers, compared to the rest
		120–150	2.1	6.9	

Bottom depth represents the deepest layer of all the sampled profiles. The error term represents standard error for $n = 3$ in Summit, $n = 4$ in Slope, $n = 3$ in Hollow, and $n = 4$ in Plain for clay content and CEC. For SSA, the error term represents standard deviation for a single sample measured three times. The table was reproduced from Berhe et al. (2012)

space that was entirely composed of grasses. The annual rate of aboveground biomass production was $444 \pm 136 \text{ g m}^{-2}$ in the Summit, $575 \pm 178 \text{ g m}^{-2}$ in the Slope, $695 \pm 202 \text{ g m}^{-2}$ in the Hollow, and $1288 \pm 701 \text{ g m}^{-2}$ in the Plain. The mean

of the landform positions ($p < 0.05$). The $\delta^{15}\text{N}$ for the Plain profile remained between 6 and 10‰ for the entire depth profile, that ranged from 2 to 200 cm to landform positions. In all values were 3% higher aboveground vegetation. Extractions of bulk soil in the all except the Plain, showed a consistent pattern with depth (Fig. 3) $\delta^{13}\text{C}$ values for bulk soil were -28 and -24‰ in all landform positions (Berhe et al. 2008). Deep soil layers in the Summit, Slope, and Plain were characterized by high N:C ratios compared to the near- surface layers. The Plain profile had an inconsistent pattern in comparison to the other landform positions, where N:C ratios remain within a narrowly constrained range of 0.08–0.1 for the entire depth profile.

N (%)	Grass	0.95 ± 0.005	0.019
	Baccharis	1.42 ± 0.033	
N:C	Grass	0.021 ± 0.0001	0.032 space
	Baccharis	0.028 ± 0.0006	
d ¹⁵ N (%)	Grass	1.99 ± 0.16	
	Baccharis	2.84 ± 0.05	
d ¹³ C (%)	Grass	-27.33 ± 0.02	-27.23
	Baccharis	-26.93 ± 0.16	-26.03
NPP (g m ⁻² year ⁻¹)		444 ± 136	

n/a represents not available, since *Baccharis* doesn't grow in the Plain. Net primary productivity (NPP) was measured by destructively harvesting aboveground

Summit Slope Hollow Plain
space

Fig. 1 a Illustration of the study hillslope and sampling locations, b N concentration of soils at the four landform positions, and c stock in the soil profiles of the eroding and

space depositional landform positions. Error bars represent SE, where $n = 4$ in Summit, $n = 8$ in slope, $n = 6$ in hollow, and $n = 9$ in plain

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Fig. 2 Rate of soil and particulate N erosion at eroding and depositional positions. Values for rates of soil erosion were determined from the literature (Heimsath et al. 1999; Yoo 2003)

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found that more than 92% of all soil nitrogen was associated with the dense fraction in all the landform positions, compared to 2–4% each that was found in the fLF and oLF (Fig. 4). We observed variable

space patterns of % N in the fLF for the different landform positions likely suggesting delivery of N into subsoil of the eroding and depositional landform positions due to episodic erosion events and/or vertical mobilization of particulate fLF down the soil profiles. We found high amount of physically protected (oLF) N around 20 cm and below 80 cm in the depositional Plain. The

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Fig. 4 Distribution of soil N in density fractions (*fLF* free light fraction, *oLF* occluded light fraction, *DF* dense fraction) space

space depositional Hollow had large proportion of N associated with soil minerals in the DF between 20–50 cm, and below 80 cm in the profile that overlapped with high proportion of N in DF in the Slope and Plain profiles around 60–80 cm. The $\delta^{15}\text{N}$ of fLF showed considerable variability within and among the different landform positions. The $\delta^{15}\text{N}$ of oLF and DF showed relatively small variability down the soil profile and was consistently higher than 2‰, compared to $\delta^{15}\text{N}$ of fLF that ranged from -1.5 to 9% (Fig. 5). In comparison, the $\delta^{13}\text{C}$ of DF was constrained between -27.5 and 25% , compared to a spread of -28 to -24% for fLF and oLF (Berhe et al. 2012).

Fig. 5 Concentration of $\delta^{15}\text{N}$ in soil density fractions (fLF free light fraction, oLF occluded light fraction, DF dense fraction) space

spacelandscape that experience lateral distribution of soil mass by erosion.

Lateral distribution of soil N by erosion

Lateral movement of soil N from the topsoil of dynamic landscapes is directly related to erosivity of rainfall (as governed by amount, distribution, and intensity of precipitation) and erodibility of soil (which depends on soil type and physical stability of aggregates), and topography of the landscapes (slope angle, slope length, curvature, and surface roughness). Our results show that soil erosion leads to spatial variability of soil N inventory at eroding versus depositional landform positions. Results presented in Fig. 2 agree with the findings of Hilton et al. (2013), where they found higher erosional losses of N (dominantly in particulate form) from steep slopes. We also observed that most of the particulate soil N that is laterally distributed by soil erosion doesn't leave the immediate watershed from which it originates, and tends to be deposited in lower-lying landform positions.

Our findings also highlight that, in addition to lateral distribution of N rich topsoil, erosion further affects soil N through its effect on soil thickness, drainage, and plant productivity. Variable drainage in eroding versus depositional landform positions is likely to exert primary control on microbial

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spaceCommunity dynamics, decomposition and composition of organic matter—including organic N containing compounds (Arnold et al. 2015; Kaiser et al. 2015). Depth profiles of soil N concentration (Fig. 1) show that soil aeration/moisture status play important roles in controlling soil N stocks in dynamic landscapes—with the well aerated Hollow storing significantly less N than the poorly-aerated Plain. Our data also shows that the depositional positions with higher soil thickness store significantly higher ($p \leq 0.05$) stock of soil N compared to the eroding landform positions with shallow soils. These patterns closely follow previous findings of higher C stocks in the poorly drained Plain, compared the well-aerated Hollow (Berhe et al. 2008).

Differential rates of accumulation and MRT of N in eroding versus depositional landform positions

To determine how fast N accumulates in soil profiles at eroding versus depositional landform positions, we used the radiocarbon concentrations in soil at each sampled soil depth. Here, we used a first-order model of N accumulation motivated by prior works on reconstructing rates of C accumulation in soil profiles (Berhe et al. 2008; Hilton et al. 2013; Jenny 1941a; Trumbore and Harden 1997), where:

$$\frac{dN_{inv}}{dt} = I - \beta \cdot N_{inv}$$

where N_{inv} is the inventory of N in the soil profile (g cm^{-2}); I is the net N input to a soil pool from all possible sources (including: atmospheric deposition,

biological N fixation, and/or deposition of eroded topsoil ($\text{g cm}^{-2} \text{ year}^{-1}$), this modeled input rate only accounts for N that is retained in soil N pool, and does not account for N that is quickly lost from a soil profile soon after it enters the system); and k is the coefficient for first-order loss of N from soil through all particulate, dissolved and gaseous pathways (year^{-1}). Equation (2) is solved subject to the initial condition $N_{\text{inv}} = N_0$ at $t = 0$ (the start of N accumulation in soil), such that:

space of the sediment during the last major landslide event in the study watershed about 13k years ago; Berhe et al. 2008, Heimsath et al. 1999; Yoo 2003), Eq. (3) can be reduced to:

$$N_{\text{inv}} = \frac{I}{k} [1 - \exp(-kt)] \quad (4)$$

For each profile, the amount of N that accumulates between successive time stages was determined by adding the N inventory of the soil layers we analyzed, starting from the bottom of the profile. For this purpose, the average age of N at each soil layer was estimated using the ^{14}C derived age of C in the same soil layer (Berhe et al. 2008). The AMS derived ^{14}C content was calibrated to derive the calendar ages using OxCal version 3.1 (Oxford University). For each profile then, the amount of N that accumulates after successive time stages is determined by adding the N inventory of each layer, starting from the bottom of the profile. After plotting cumulative N inventory versus years of accumulation (t), we determined the values of I and k by fitting the data with Eq. (4) using the Solver function in Excel (MS Office 2011). This method provides a first rate approximation of the rate of N accumulation and residence time in these soil profiles since: (a) most N in soil is associated with SOM (Matheny and Hunt 1983); (b) more than 95% of N stored in soils is found in organic forms, and most organic N in soil is covalently bonded to carbon-based organic compounds in soil (Allison 1973; Batjes 1996); and (c) for all our samples and locations, the concentration of C and N are very strongly correlated (R^2 values of ≥ 0.97).

Results from the above formulation show that rate of soil N accumulation is distinctly different in eroding versus depositional landform positions. The eroding landform position soil profiles (Summit and Slope) reached state N accumulation about 600

and 2000 years after the start of the model spin up. By comparison, the soil profiles in the two depositional landform positions (Hollow and Plain) appear to be approaching steady state only after 6000 years of model spin up, at least 4000 years after the eroding

$$I = \frac{dN_{\text{inv}}}{dt} + kN_{\text{inv}}$$

space positions (Fig. 6). From the model runs, we derived an

$$N_{\text{inv}} = \frac{I}{k} [1 - \exp(-kt)]$$

estimate of the profile integrated mean residence time

of N: 189 years in the Summit, 1198 years in the Slope. Noting that $N_0 = 0$ for both the eroding hillslopes (at the time of first soil development) and the depositional basins (assuming complete evacuation of N), we derived an estimate of the profile integrated mean residence time of N: 3795 years in the Hollow, and 2106 years in the Plain. The modeled rate of N input into the soil N pool in the Summit was 14-times higher than the rate of N

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Fig. 6 Rate of N accumulation in the different landform positions. Total amount of N accumulated versus mean age of soil at the depth above which N is inventoried. “Years of accumulation” were derived from calibrated radiocarbon ages of SOM at the points of measurement (see “Methods” section), where fits are based on ^{14}C ages of bulk soils; $t = 0$ at time of soil formation in slopes, and at start of C accumulation, after landslide (13 k years ago) in depositional sites. Method based on Berhe et al. (2008) and Trumbore and Harden (1997)

erosion, compared to 4-times higher rate of N input compared to erosion in the Slope. In the depositional landform positions, the modelled rate of N input into the soil pool was 5-times and 8-times higher than the rate of N input through deposition of eroded soil material in the Hollow and Plain, respectively. The erosional loss of soil N is equivalent to 7 and 25% of the modeled N input in the eroding landform positions of Summit and Slope, respectively (Table 3).

Controls on N storage and persistence in dynamic landscapes

Soil moisture gradients that are associated with topography tend to have important effects on N input, transformations, and loss from the soil system (Berhe

space2013; Stewart et al. 2014). Typically, high fluxes of gaseous (as nitrous oxide, N_2O) and dissolved (as organic or inorganic N forms) N species from soils are observed from lower-lying and gently sloping landform positions that are usually characterized high soil water content and high organic matter stock (as in the Plain),

compared to well-aerated soils (as in the upper parts of the catchment in our site) (Brady and Weil

2008). However, despite the likelihood of high N loss rate from the depositional plain, in our study site, we observed highest stock of soil N in the depositional plain. In fact, the stock of total N was

two to three times higher in the poorly drained depositional Plain compared to the eroding landform positions (Table 2). It is likely then that

the stock of N in the Plain is maintained by continuous input of N from deposition of eroded topsoil, in situ recycling of soil N by plants in the Plain, and potential contribution from biological N fixation. Our results show that over 90% of the soil N is associated with the dense fraction (DF),

where N containing functional groups form chemical association with soil mineral surfaces.

This result agrees with past studies that showed that over 90% of soil N is likely associated with

“recalcitrant” OM in soil (Schlesinger 1997). The conceptual basis of OM recalcitrance is challenged in biogeochemistry today (Kleber 2010) because of

overwhelming evidence showing that OM persistence is a function of ecosystem variables and not its biochemical composition, which was previously thought to render it “recalcitrant”

(Schmidt et al. 2011). Nonetheless, we can assume that OM with long residence time in soils most likely resides in the protected pools (oLF and DF in our density fractionation scheme), and that the majority of all the N soil is indeed associated with this relatively stable pool of N (compared to the fLF and dissolved

pools of N in soil).

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spaceTable 3 Soil N inventory (N_{inv}) and rate of soil N erosion (N_e , positive) or deposition (N_d , negative), and modeled values of whole soil profile-integrated net input of N to the soil N pool

space(I_N), modeled coefficient for first-order N loss from for the soil profile (k), and modeled mean residence time of N (MRT_N) in soil profiles at each of the four landform positions

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Location	N_{inv} (g cm ⁻²)	N_e (?) or N_d (-) (g cm ⁻² year ⁻¹)
Summit	0.14	4.69E-05
Slope	0.18	2.57E-05
Hollow	0.30	-1.78E-05
Plain	0.42	-2.37E-05

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spaceChemical association of N with reactive soil minerals

To determine how soil N storage is related to concentration of reactive minerals, we refer to previously published data from three selective dissolutions for Fe and Al oxides, hydroxides and oxyhydroxides (of the form MO_x , $M(OH)_x$, or MO_xOH_x respectively, where M is the metal (Fe and Al)), that were performed to determine the stock of three classes of mineral functional groups that can chemically protect organic compounds from decomposition—crystalline Fe, poorly crystalline Fe and Al oxides, and chelated organo-metal complexes (Berhe et al. 2012). We found a statistically significant positive linear association of N concentration with pyrophosphate extractable Fe and Al (Fe_{py} and Al_{py} , Table 3) in the Summit, Slope and Plain positions. The strong association of soil N concentration with pyrophosphate extractable Fe and Al suggests that most of the N stabilized by association with reactive soil minerals is likely to be in the form of N containing organic compounds that form complexes with this class of reactive minerals. The variability of poorly crystalline Fe (Fe_{ox-py}) concentrations in the soil only explained a significant portion of the variability of soil N concentration in the Summit position. The concentrations of poorly crystalline Fe and Al (oxalate extractable) or total pedogenic Fe (dithionite extractable) did not have significant linear association

with N concentration in the soils at any of the landform positions. The linear association of N concentration with concentration of pyrophosphate and oxalate extractable Fe and Al was strongest in the Summit position, characterized by higher rates of soil production. Previous work in the study area also found a statistically significant linear correlation between C concentration and the concentrations of Fe and Al oxy(hydr)oxides (Berhe et al. 2012), suggesting a strong coupling of both C and N persistence in soil through association with reactive minerals. The influence of metal oxides on N storage was stronger in eroding positions, where we found R^2 between 0.60 to 0.83 for the association of N with Al_{py} , Fe_{py} , and Fe_{ox-py} in Slope and Summit. In comparison, for the depositional position soil profiles, the only significant association of N storage with metal oxides that we found was $R^2 = 0.79$ for association of N

spaceoxides in the Hollow (Table 4). This major difference in association of metal oxides in the well-drained Hollow versus the poorly-drained Plain suggest that hydrology is likely to be an important control on N storage and loss in such dynamic landscapes (Stewart et al. 2014). Findings of high N:C ratio at depth in Summit, Slope, and Plain, along with increased ^{14}C age in these deep soils (Berhe et al. 2008), suggests more efficient retention of N compared to C, consistent with consumption and cycling of organic compounds by heterotrophic organisms (Hilton et al. 2013).

Transformations of soil N in eroding versus depositional landforms

The stable isotope composition of N is more useful for determining N dynamics than bulk elemental concentration, because of its ability to integrate ecological processes over both spatial and temporal scales (Robinson 2001). The stable isotope ratio of N in soils represents an integration of in situ biological fixation and recycling of N in soil, decomposition/ mineralization of organic matter; and gaseous, dissolved, and particulate flows among different N reservoirs in the earth system. Previously studies have demonstrated the utility of $d^{15}N$ a useful proxy to determine impact of climate on soil N dynamics (Natelhoffer and Fry 1988), and the role of soil physical properties such as clay content on N dynamics (Craine et al. 2015). Furthermore, $d^{15}N$ is also a useful tool in

determining prominent loss pathways of N in soil. It is accepted that soils with high $d^{15}N$ likely lose more N through NH_3 volatilization and denitrification (strongly ^{15}N discriminating loss pathways) compared to soils with low ^{15}N that are more likely to lose most of their N through less ^{15}N discriminating pathways such as dissolved N (DON or NO_3^-)

R, %N with	Summit	Slope	Hollow	Plain
%C	0.99	0.99	0.99	1.00
Al_{py}	0.83	0.70		0.79
Fe_{py}	0.90	0.60		
$SPFe_{ox-py}$	0.65			

no significant association of N storage with metal

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spaceleaching and subsurface flow processes (Craine et al. 2015). Moreover, Perakis et al. (2015) showed that, in montane ecosystems on steep topography, a decline in soil $d^{15}N$ values suggests loss of N by soil erosion and/ or rapid cycling of N in soil. In our study site, the significantly higher $d^{15}N$ of bulk soil in the Plain suggests that the poorly drained Plain landform position site likely loses considerable proportion of soil N through denitrification. This conclusion is consistent with our observations that the Plain site can be waterlogged for months, where N transformations take place under anaerobic conditions that would be conducive for denitrification.

The pattern of $d^{15}N$ with depth for both the bulk soil (Fig. 3) and the different density fractions (fLF, oLF, and DF, Fig. 5) deviate from the usually observed patterns in soil, where $d^{15}N$ values are negative in topsoil and get progressively more positive down the soil profile (Hobbie and Ouimette 2009). In dynamic landscapes, the deviation from the usually observed pattern with depth is likely a reflection of the physical mixing of the soil (due to burrowing mammals) and profile inversion associated with soil erosion where material that used to be in topsoils of eroding landform positions gets buried deep in the profile of depositional landform positions after successive erosion and deposition events (Berhe et al. 2007). In our study site, previous studies had noted considerable mixing of the soil by pocket gophers (Berhe et al. 2008; Yoo et al. 2005b), and burial of soil due to the processes of soil erosion and deposition (Berhe et al. 2007, 2014). In particular, the $d^{15}N$ of fLF gets progressively lower, or less positive with depth suggesting that OM in fLF in topsoil layers of the depositional plain is likely more processed than fLF in deep soil layers owing to rapid

burial and preservation of relatively less decomposed fLF at depth. On the other hand, we don't see low $\delta^{15}\text{N}$ of fLF in near-surface layers in the Plain presumably because fLF that remains in topsoil layers would be exposed to more chemical transformation associated with decomposition, compared to fLF in deep soil layers of the same landform position. This pattern of decreasing $\delta^{15}\text{N}$ of fLF with depth is unique to the depositional Plain, likely suggesting that fLF buried in deep soil layers of poorly drained depositional landform positions is likely to be preserved more efficiently than fLF buried in deep soil layers of well-drained soil profiles.

In addition to physical mixing of soil due to activity of burrowing mammals and erosion, the results in Figs. 3 and 5 also suggest intense rates of N processing and SOM transformations in the soil profiles of this dynamic landscape. The $\sim 4\%$ $\delta^{15}\text{N}$ of fLF in the topsoil layers of all landform positions and the entire profile of the Hollow suggest intense microbial transformation of SOM (Meyer and Leifeld 2013) supporting the idea that SOM in these landform positions and soil layers is more processed than one would expect for relatively new SOM in topsoil. This is plausible considering the above reported rates of topsoil erosion in this catchment, where every landscape position other than the Summit is likely to receive and at least temporarily store N that was eroded from the landform positions that lie above it. Furthermore, every landform position is also likely to receive N input from atmospheric deposition (which we were not able to account for in this study).

Relating N dynamics with decomposition related transformation of SOM composition

Detailed investigation into the stable and isotopic compositions of N can shed light on how certain

storage and stabilization in the soil system. Kramer et al. (2003) showed that a strong positive correlation between the degree of decomposition of plant material expressed as the alkyl/O-alkyl ratio (from CP MAS ^{13}C -NMR) and $\delta^{15}\text{N}$ can be used as an indication that microbial processing of SOM is a stronger control on changes in SOM composition, rather than selective preservation of organic compounds that are highly resistant to decomposition. To investigate how N dynamics in soil is related to SOM composition, we refer to previously published NMR data from the same study site. In Berhe et al. (2012) composition of SOM and biochemical changes associated with decomposition were determined using solid state cross polarization (CP) magic angle spinning (MAS) ^{13}C -nuclear magnetic resonance (NMR) (Baldock et al. 1992, 1997). Here we use the relative difference in proportion of total OM in the alkyl and O-substituted alkyl regions in the ^{13}C -NMR spectra [i.e. alkyl to O-alkyl ratio (A:O-A)] as an index of the degree of decomposition in soil organic compounds (Baldock et al. 1997). We found a strong positive correlation

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between $\delta^{15}\text{N}$ and the alkyl/O-alkyl ratio (of bulk soil and DF) only in the Plain position (Fig. 7), suggesting that the extent of N storage and stabilization by sorptive interaction with minerals is controlled by microbial processing of organic substrates only in the poorly drained depositional Plain. We found negative correlation between $\delta^{15}\text{N}$ and the alkyl/O-alkyl ratio in the bulk SOM of Summit, Slope, and Hollow, as well as the fLF and oLF of both Slope and Plain positions, suggesting that factors other than microbial alteration of organic matter are likely responsible for changes in SOM composition in such dynamic landscapes. In hillslope soil profiles characterized by significant lateral surface and subsurface flow of water, accumulation of organic functional groups that are left behind after leaching, and lateral mobilization of dissolved and particulate organic compounds is likely to have a significant effect on the composition of bulk SOM. Similarly, even though this was not addressed in our study, transformation of plant biomass during burning can also have important implications for both storage and stabilization of SOM (Rumpel et al. 2006a).

It is likely that the SOM in the different soil layers and fractions at such dynamic landscapes undergoes periodic stabilization and destabilization that is facilitated by variable wetting and drying cycles and

Fig. 7 Relating $\delta^{15}\text{N}$ with NMR derived index of SOM transformation

spaceexperience considerable physical perturbations due to mixing and lateral redistribution of soil due to soil erosion.

Conclusions

Geomorphology exerts a strong control on the storage of soil N and its distribution within the soil matrix and profile. We found that erosion and its effects on rates of soil development, particle size distribution, soil mineralogy, soil thickness (depth), soil organic matter concentration, and hydrology at different landform positions in the landscape (Berhe and Kleber 2013) exerts significant influence on the storage and loss of N from eroding watersheds. The findings of this study emphasize the local as well as global significance of erosion as an important mechanism that controls N dynamics. Considering the anticipated changes in a future climate that include intensification of the hydrologic cycle, it is also plausible to expect higher rates particulate N flux from the terrestrial ecosystem (Hilton et al. 2013), with multiple implications for ecology and biogeochemistry of aquatic ecosystems. Further studies are needed to determine spatial distribution of N in other types of dynamic landscapes, and how lateral distribution not just by water, but also by wind erosion, controls N storage in the soil system.

Acknowledgements We thank John Harte and Jennifer Harden for their helpful comments during the research process and comments on earlier versions of this manuscript. Funding for this work was provided by the National Research Initiative Competitive Grant 2003-35107-13601 from the USDA Cooperative State Research, Education, and Extension Service, National Science Foundation (CAREER, EAR- 1352627), and the U.S. Department of Energy, Office of Science, [Office of Biological and Environmental

Research, Atmospheric System Research Program] under Award Number DE-AC02-05CH11231. The authors have declared that no competing interests exist.

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$$N_{inv} = \sum \Delta Z_i \cdot \rho_i \cdot (1 - R_i) \cdot (\%N \cdot 100)$$

community dynamics, decomposition and turnover of organic matter—including organic acids and amino compounds (Arnold et al. 2015; Kaise et al. 2015). Depth profiles of soil N concentration (Fig. 2) indicate that soil aeration/moisture status play important roles in controlling soil N stocks in dynamic landscapes. The well aerated Hollow storing significantly more N than the poorly-aerated Plain. Our data indicate that the depositional positions with high elevation store significantly higher ($p < 0.05$) soil N compared to the eroding landform positions and the shallow soils. These patterns closely follow the findings of higher C stocks in the poorly aerated soils compared the well-aerated Hollow (Berhe et al. 2008).

Differential rates of accumulation and loss of N in eroding versus depositional landform positions

To determine how fast N accumulates in soils in eroding versus depositional landform positions, we used the radiocarbon concentrations in soil profiles sampled soil depth. Here, we used a first-order model of N accumulation motivated by prior work in reconstructing rates of C accumulation in soils (Berhe et al. 2008; Hilton et al. 2013; Jandl et al. 2007; Trumbore and Harden 1997), where:

$$\frac{dN_{inv}}{dt} = I - k \cdot N_{inv}$$

where N_{inv} is the inventory of N in the soil (g cm^{-2}); I is the net N input to a soil profile from all possible sources (including: atmospheric N deposition, biological N fixation, and/or deposition of N in topsoil ($\text{g cm}^{-2} \text{ year}^{-1}$), this modeled in the model accounts for N that is retained in soil N pools and does not account for N that is quickly lost from the system soon after it enters the system); and k is the first-order loss rate for first-order loss of N from soil through