



Decoupling of sulfur and nitrogen cycling due to biotic processes in a tropical rainforest

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Abstract

We examined the terrestrial sulfur (S) cycle in the wet tropical Luquillo Experimental Forest (LEF), Puerto Rico. In two previously instrumented watersheds (Icacos and Bisley), chemical and isotopic measurements of carbon (C), nitrogen (N) and S were used to explore the inputs, in-soil processing, and losses of S through comparison to the N cycle. Additionally, the impact of soil forming factors (particularly climate, organisms, topography and parent material) on S cycling in this system was considered. Atmospheric inputs ($\delta^{34}\text{S}$ values of $16.1 \pm 2.8\text{‰}$), from a mixture of marine and anthropogenic sources, delivered an estimated $2.2 \text{ g S}/(\text{m}^2\text{yr})$ at Icacos, and $1.8 \text{ g S}/(\text{m}^2\text{yr})$ at Bisley. Bedrock N and S inputs to soil were minimal. We estimated a hydrologic export of $1.7 \pm 0.1 \text{ g S}/(\text{m}^2\text{yr})$ at Icacos, and $2.5 \pm 0.2 \text{ g S}/(\text{m}^2\text{yr})$ at Bisley. Stream baseflow S isotope data revealed significant bedrock S in the hydrologic export at Bisley (with a distinctive $\delta^{34}\text{S}$ values of $1.6 \pm 0.7\text{‰}$), but not at Icacos. Pore water data supported the co-occurrence of at least three major biological S-fractionating processes in these soils: plant uptake, oxidative degradation of organic S and bacterial sulfate reduction. The rates and relative importance of these processes varied in time and space. Vegetation litter was 3–5‰ depleted in ^{34}S compared to the average pore water, providing evidence for fractionation during uptake and assimilation. Out of all abiotic soil forming factors, climate, especially the high rainfall, was the main driver of S biogeochemistry in the LEF by dictating the types and rates of processes. Topography appeared to impact S cycling by influencing redox conditions: C, N and S content decrease downslope at all sites, and the Bisley lower slope showed strongest evidence of bacterial sulfate reduction. Parent material type did not impact the soil S cycle significantly. To compare the fate of S and N in the soil, we used an advection model to describe the isotopic fractionation of total S and N associated with downward movement of organic matter in both dissolved and solid fractions. This model worked well for N, but the assumption of a constant fractionation factor α with depth failed to describe S transformations. This result revealed a fundamental difference between N and S cycling in these soils, indicating an apparent greater sensitivity of S isotopes to fluctuating redox conditions.

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Abbreviations: COS, carbonyl sulfide; CZO, Critical Zone Observatory; DMS, dimethyl sulfide; DOS, dissolved organic sulfur; LEF, Luquillo Experimental Forest; LTER, Long-Term Ecological Research; MAP, mean annual precipitation; masl, meters above sea level; MAT, mean annual temperature; NAD83, North American Datum of 1983; NRCS, National Resources Conservation Service; nss, non-seasalt; SOS, soil organic sulfur; USDA, United States Department of Agriculture; USGS, United States Geologic Survey

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1. INTRODUCTION

The terrestrial biogeochemical cycle of an element is driven by inputs, outputs, and transformations within the plant/soil system (e.g. Vitousek and Sanford, 1986). Sulfur (S), like nitrogen (N), has a complex biogeochemical cycle for two main reasons. First, S exists in a wide range of valence states (from -2 to $+6$), and can participate in intricate biochemical reactions, some of which are challenging to elucidate (Norman et al., 2002; Brunner and Bernasconi, 2005; Bradley et al., 2011; Sim et al., 2011). Second, S is an essential nutrient, but soils can suffer both from S deficiency (Tabatabai, 1984; Acquaye and Beringer, 1989), and from S excess (via pollution, or naturally sulfide-rich bedrocks) (Likens et al., 2002). Unlike N however, for which both the natural and the human-impacted cycles have been well studied, S has been studied mostly in industrialized regions subject to high deposition rates (e.g. Novak et al., 2001; Likens et al., 2002; Marty et al., 2011), and we still lack a basic understanding of the natural controls on the S cycle in less perturbed environments. Based on N studies (e.g. Austin and Vitousek, 1998; Amundson et al., 2003), it is likely that the soil formation factors identified by Jenny (1941) (climate, organisms, topography, parent material and time) play a role in the soil S cycle, however no previous research has addressed these controls systematically.

A potential impact of topography was observed in Costa Rica, where Bern and Townsend (2008) found that hillslope soils had higher $\delta^{34}\text{S}$ values compared to alluvial soils. Climate should have a first order impact on redox-sensitive elements, as mean annual precipitation (MAP) impacts elemental input and loss rates, and the dominant biological processes in the soil, while mean annual temperature (MAT) impacts the rates of biotic processes such as bacterial sulfate reduction (Bruchert et al., 2001; Canfield et al., 2006; Turchyn et al., 2010). Previous research found that wet tropical forest soils may emit hydrogen sulfide due to dissimilatory sulfate reduction (Delmas et al., 1978; Delmas and Servant, 1983; Newman et al., 1991), however this was shown not to be an important process in Costa Rica, where most of the S research on pristine wet tropical forest soils to date has focused (Bern et al., 2007; Bern and Townsend, 2008). Hillslope soils in Costa Rica depend exclusively on atmospheric S inputs, show minimal variation in concentration and isotope values with depth, and an overall slight enrichment in ^{34}S compared to the precipitation. This isotopic enrichment may be due to emissions of highly depleted biogenic S gases by vegetation, or due to oxidative degradation of organic S (Bern et al., 2007; Bern and Townsend, 2008). Fractionation during plant uptake is commonly assumed to be negligible due to similar total soil and plant isotope values (Bern and Townsend, 2008). The largest S isotope fractionations observed in nature occur during microbial dissimilatory sulfate reduction, which can deplete the sulfide products by up to 70‰ (Brunner and Bernasconi, 2005). Despite their generally small magnitude (Table A1), other biological fractionations sensitive to climate are also significant because they all operate in the same direction: microbes and plants

preferentially utilize the lighter isotope during metabolism, therefore $\delta^{34}\text{S}$ values tend to decrease in products compared to the substrates (see Table A2 for a compilation of input $\delta^{34}\text{S}$ values). An overview of the soil S cycle, based on current knowledge, is illustrated in Fig. A1.

Here we use stable isotopes to investigate the geochemical cycling of S in the wet montane forest ecosystem of the Luquillo Experimental Forest (LEF) in Puerto Rico, and compare it to the better-known N cycle. At the LEF, previous studies have focused on rates of chemical weathering and solute loss (e.g. White et al., 1998; Murphy et al., 1998; Schulz and White, 1999; Buss et al., 2008), and on the sources and cycling of nutrients such as N and P (e.g. Silver et al., 1994; Pett-Ridge et al., 2009; Buss et al., 2010). Sulfate and/or total S concentrations have been measured in precipitation (McDowell et al., 1990; Asbury et al., 1994; White et al., 1998; Heartsill-Scalley et al., 2007), soil and saprolite (Stanko-Golden and Fitzgerald, 1991; White et al., 1998; Cox et al., 2002), and major streams (McDowell and Asbury, 1994; White et al., 1998; Bhatt and McDowell, 2007). Here we integrate and expand on these previous studies and present novel stable S and N isotope data for the coupled precipitation-soil-vegetation system, to address two yet unanswered questions: (1) how do soil forming factors, particularly bedrock and topography, impact S biogeochemistry in a tropical environment? and (2) are the N and S biogeochemical cycles coupled, or at least similar, in tropical soils? These questions have importance for understanding spatial patterns of nutrient dynamics and feedbacks, and for setting a baseline for studying the response of tropical soils to climate or land use change.

2. SITE DESCRIPTION

The Luquillo Experimental Forest (LEF) ($18^{\circ}18'\text{N}$, $65^{\circ}50'\text{W}$, referenced to the NAD83 datum) is a Long-Term Ecological Research (LTER) and Critical Zone Observatory (CZO) site, with a long history of biogeochemical research (e.g. Scatena, 1989; McDowell et al., 1990; Silver et al., 1994; White et al., 1998). Mean annual precipitation (MAP) in this warm and humid tropical forest increases with elevation, ranging from about 2500 mm to over 5000 mm at the highest elevation of 1074 m (Scatena, 1989; McDowell and Asbury, 1994). Rainfall is significant year round, with January through April being the driest period (Heartsill-Scalley et al., 2007). Precipitation occurs mostly as frequent, short, high intensity events (Scatena, 1989; White et al., 1998; Buss et al., 2010). Convective boundary layer storms with strong orographic effects are most common (White et al., 1998), but northeasterly trade winds, winter cold fronts, tropical storms, depressions and hurricanes also affect the region (Heartsill-Scalley et al., 2007). Mean annual temperature (MAT) is $22\text{--}23^{\circ}\text{C}$ at both sites (White et al., 1998; Murphy and Stallard, 2012).

Our study focused on two LEF watersheds instrumented by the USGS (White et al., 1998; Buss et al., 2011; Buss and White, 2012): Icacos (mature Colorado forest on quartz diorite) and Bisley (mature Tabonuco forest on volcanoclastics) (Fig. 1). The Icacos site is located on the shoulder of the Guaba ridge in the catchment of the Guaba stream, a

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