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Linking soil element-mass-transfer to microscale mineral weathering across a semiarid environmental gradient

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ABSTRACT

Understanding controls on silicate weathering is critical to characterizing critical zone evolution. The objective of this study was to investigate how climate, vegetation, and landscape position control feldspar transformations across a semiarid environmental gradient. Granitic surface soil and saprock samples were collected from desert scrub and mixed conifer sites within the Santa Catalina Mountain Critical Zone Observatory where mean annual temperature ranges from 24 °C to 10 °C and mean annual precipitation from 25 to 85 cm. Quantitative X-ray diffraction, X-ray fluorescence, and electron microprobe analyses were employed to quantify elemental changes in bulk soils and across plagioclase grains. The chemical depletion of Na in bulk soils ranged from 5.4 – 15% in the desert scrub sites relative to 16–33% in the mixed conifer sites. Plagioclase grain alteration was classified into unaltered, edge, and altered sections to compare microscale weathering and elemental variation. The Na/Al and Si/Al ratios decreased from unaltered, to edge, to altered grain sections in the mixed conifer sites, whereas the element ratios of the desert scrub system were similar between unaltered and edge grain sections, and only exhibited significant decreases in Na/Al and Si/Al ratios between edge and altered materials. The microscale depletion of Na and Si suggested increased silicate weathering in the cooler, wetter, and more biologically productive mixed conifer system compared to the hot, dry desert scrub system. The results also demonstrated a topographic control on mineral transformation where increased plagioclase weathering occurred in convergent footslope landscapes with little change in elemental depletion of soils in divergent summit sites.

1. Introduction

Silicate weathering and mineral transformations contribute significantly to landscape evolution and to our understanding of water and carbon cycling in the critical zone, which spans from the top of the vegetation canopy down to and including groundwater (NRC, 2001). Important questions remain regarding the role of various climatic, tectonic, and topographic factors in controlling silicate weathering mechanisms and microscale transformations in the critical zone. Here, we address how climate, vegetation, and landscape position interact to shape silicate weathering patterns through elemental loss in bulk soils and through microscale transformations of plagioclase feldspar grains.

The dominant controls on silicate weathering and the nature of the subsurface weathering front include water availability and temperature (White and Brantley, 1995; White et al., 1996; Dahlgren et al., 1997; White et al., 2001; Rasmussen and Tabor, 2007; Rasmussen et al., 2010), physical erosion (Riebe et al., 2001, 2004), and the interactions between climate and erosion (Jacobson et al., 2003; West et al., 2005; Rasmussen et al., 2011). Additional factors reported to control extent and rates of silicate weathering include deep regolith and saprolite weathering (Dixon et al., 2009), vascular plants and primary production (Moulton et al., 2000), and topographic controls on soil production rates, mineral residence time, soil and solute transport distances, and soil depth (Heimsath et al., 1997; Green et al., 2006; Yoo et al., 2007; Yoo and Mudd, 2008). At the mineral scale, White and Brantley (2003) suggest that silicate weathering reaction rates are a product of both extrinsic properties, such as solute composition, climate, and/or biologic interactions, and intrinsic properties. Intrinsic properties can include increases in grain surface roughness and concurrent increases in mineral surface area or decreases in reactive surface area due to physical impediment by secondary weathering products or leached layers. Nearly all of these factors co-vary and interact, and thus present a challenge to understanding chemical weathering processes and critical zone evolution (Chorover et al., 2011).

The microscopic study of weathered mineral surfaces in the critical zone may be used to constrain the formation and distribution of secondary altered products, an aspect not readily addressed in laboratory investigations of feldspar weathering (Hochella and Banfield, 1995). Of the silicate minerals, feldspars comprise approximately 60% of the minerals in the Earth's crust (Kauffman and Van Dyk, 1994) and have been studied extensively in laboratory weathering experiments (e.g., Blum

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and Stillings, 1995). However, feldspar dissolution rates are consistently two to five orders of magnitude higher in laboratory settings compared to field studies (Blum and Stillings, 1995; White et al., 1996, 2001; White and Brantley, 2003; Zhu et al., 2006), likely a result of differences in mineral surface characteristics of freshly ground and naturally weathered feldspars. High-resolution imaging and accompanying chemical analyses present a powerful tool to investigate silicate weathering processes in critical zone systems that may help address this discrepancy, particularly in regard to the composition and mineral transformations that occur at reactive feldspar surfaces. Zhu et al. (2006) illustrated the ambiguous definitions associated with the feldspar surface–water interface that include but are not limited to: reactivity dominated by surface reactions between unaltered feldspar and the aqueous solution (Lagache, 1961); the occurrence of a cation deficient leached layer separating the reactive feldspar surface from aqueous solution (Paces, 1973); mineral dissolution controlled by non-diffusive, chemical reactions at the feldspar surface–water interface (Berner and Holdren, 1977, 1979); the existence of a sharp feldspar–amorphous silica chemical gradient created by interfacial dissolution–reprecipitation processes (Hellmann et al., 2003, 2012); or a complex surface where crystalline feldspar is separated from the aqueous solution by a thin (<10 nm) amorphous layer covered by discontinuous kaolinite and an outer layer of precipitated smectite. Similar to White and Brantley (2003), Zhu et al. (2006) related differences in feldspar reactivity of laboratory and field samples to intrinsic mineral properties including amorphous layer development and secondary mineral coatings on feldspars.

The objective of this work was to constrain how climate and vegetation, topographic position, and pedon depth control plagioclase weathering across a steep semiarid environmental gradient. Local environmental gradients that compress large climate and vegetation variation over short distances provide excellent opportunities to examine the interaction among these factors with minimal variation in bedrock and regional tectonics (Dahlgren et al., 1997). Plagioclase feldspar chemical composition in bulk soils was coupled with microscale elemental changes associated with feldspar chemical transformation. Electron microprobe wave dispersive spectroscopy (WDS) combined with backscattered electron (BSE) imagery was used to develop a classification scheme for characterizing the plagioclase grain–secondary mineral interface.

2. Methods

2.1. Field sites and sample collection

The two field sites, referred to here as *desert scrub* and *mixed conifer*, were sampled from the Santa Catalina Critical Zone Observatory environmental gradient in southern Arizona (Fig. 1a). These locations capture the relative end-members of the environmental gradient and encompass the largest variation in climate, vegetation, and topography. The Santa Catalina Mountains encompass significant range in temperature (10–24 °C) and precipitation (25–85 cm), with concurrent variation in vegetation community composition and structure (Whittaker and Niering, 1965; Whittaker et al., 1968; Table 1). Climate and vegetation both exert control on soil properties and silicate weathering, with distinct variation in soil development by vegetation community, or climate–vegetation zone (Whittaker et al., 1968). Additionally, local relief and topography vary significantly across the SCM (Pelletier et al., 2013), leading to landscape position variation in soil physical and chemical properties within each climate–vegetation zone (Lybrand et al., 2011).

A 1:125,000 digital spatial database containing geologic maps of the Catalina Core Complex and field observations of rocks were used to determine the geology of the field sites, which span two Tertiary-aged intrusive rock units (Dickinson, 1991, 2002). Specifically, the desert scrub site was located on the Oligocene–Miocene aged Catalina granitic pluton, and the mixed conifer site was located on the Eocene aged two-mica wilderness granite suite. The granitic parent materials exhibit similar mineral and elemental composition except for the dominant mica mineral where the desert scrub site is dominated by biotite and the mixed conifer location is dominated by muscovite (Table 2).

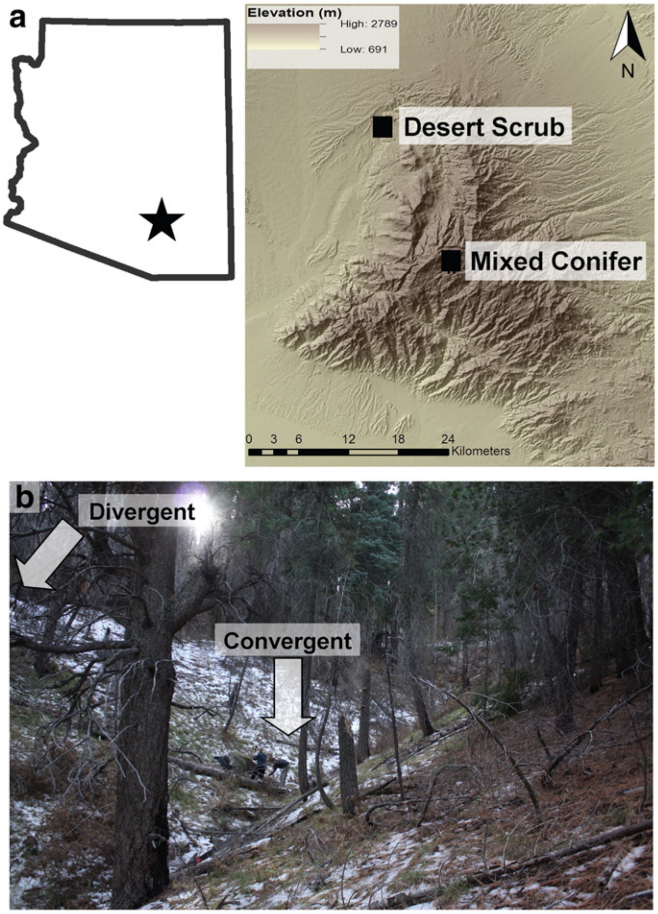

Fig. 1. a) Locations of the desert scrub and mixed conifer sites encompassed by the Santa Catalina Mountain environmental gradient in southern Arizona. b) An example of the two landscape positions studied in the project. The image is of an adjacent divergent–convergent landform unit pair at the mixed conifer field site.

Local topographic controls on silicate weathering were examined in the desert scrub and mixed conifer systems by sampling a combination of divergent summit landscape positions and adjacent convergent footslope positions. The divergent summit positions are a source of soil materials and solutes to adjacent convergent footslope sites where increased water availability and colluvial inputs likely enhance silicate weathering and mineral transformation (Huggett, 1975; Birkeland, 1999). The sample location design follows similar hillslope scale soil studies that focus on the interactive control of climate and topography on granitic soil development (Watson, 1964; Muhs, 1982; Khomo et al., 2011).

Soil, saprock, and representative parent rock samples were collected from the desert scrub and mixed conifer locations (Fig. 2a, b). Pedons were excavated to the depth of refusal from north-facing convergent and divergent landscape positions for a total of four pedons per sample location. Pedon locations were selected based on field observations of local topography and landscape configuration. Soils and saprock were sampled by morphologic horizon and described using established methods (Schoeneberger et al., 2011). Soil subgroup classifications and dominant vegetation were also determined for each field site (Table 1). The weathered parent material observed at the sites in the current study best matched the characteristics of saprock as outlined by Graham et al. (2010), where saprock is defined as weathered parent material that retains the structural features of the parent rock, can be broken apart with bare hands, and is dominated by primary minerals