



## Strong climate and tectonic control on plagioclase weathering in granitic terrain

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### ABSTRACT

Investigations to understand linkages among climate, erosion and weathering are central to quantifying landscape evolution. We approach these linkages through synthesis of regolith data for granitic terrain compiled with respect to climate, geochemistry, and denudation rates for low sloping upland profiles. Focusing on Na as a proxy for plagioclase weathering, we quantified regolith Na depletion, Na mass loss, and the relative partitioning of denudation to physical and chemical contributions. The depth and magnitude of regolith Na depletion increased continuously with increasing water availability, except for locations with mean annual temperature  $<5\text{ }^{\circ}\text{C}$  that exhibited little Na depletion, and locations with physical erosion rates  $<20\text{ g m}^{-2}\text{ yr}^{-1}$  that exhibited deep and complete regolith Na depletion. Surface Na depletion also tended to decrease with increasing physical erosion. Depth-integrated Na mass loss and regolith depth were both three orders of magnitude greater in the fully depleted, low erosion rate sites relative to other locations. These locations exhibited strong erosion-limitation of Na chemical weathering rates based on correlation of Na chemical weathering rate to total Na denudation. Sodium weathering rates in cool locations with positive annual water balance were strongly correlated to total Na denudation and precipitation, and exhibited an average apparent activation energy ( $E_a$ ) of  $69\text{ kJ mol}^{-1}$  Na. The remaining water-limited locations exhibited kinetic limitation of Na weathering rates with an  $E_a$  of  $136\text{ kJ mol}^{-1}$  Na, roughly equivalent to the sum of laboratory measures of  $E_a$  and dissolution reaction enthalpy for albite. Water availability is suggested as the dominant factor limiting rate kinetics in the water-limited systems. Together, these data demonstrate marked transitions and nonlinearity in how climate and tectonics correlate to plagioclase chemical weathering and Na mass loss.

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### 1. Introduction

*What controls the thickness and chemistry of the Earth's regolith?* Answering this question is fundamental to understanding landscape evolution, soil-forming processes, and the feedback between mineral weathering, the global cycling of chemical elements, and atmospheric carbon dioxide (Bernier et al., 1983; Raymo and Ruddiman, 1992). On upland landscapes, the chemistry and thickness of regolith, defined here to include soil and saprolite layers down to the depth of unweathered bedrock, are thought to be controlled by parent material mineralogy, precipitation, temperature, physical erosion, biota, and dust input (Dokuchaev, 1967; Jenny, 1941). The lack of models that can predict the thickness and chemistry of regolith as a function of these variables is due to difficulties in deciphering the relative contribution of each variable, as well as observed thresholds in soil

development (Chadwick and Chorover, 2001), landscape evolution (Phillips, 2006), and mineral weathering (Kump et al., 2000).

Climate is considered a central control on regolith formation, manifest as water availability and temperature (White, 1995). Water causes silicate hydrolysis and transports soluble weathering products away from reaction sites, whereas temperature controls weathering rates and mineral solubility (Lasaga et al., 1994). Temperature also moderates water availability through its control on the partitioning of precipitation to evapotranspiration and base-flow (L'vovich, 1979). Variation in climate is coupled with concomitant variation in biota and biotic influences on chemical weathering via transfer of  $\text{CO}_2$  and reduced carbon compounds into the regolith. However, the direct role of biota can be very difficult to separate from climate because of their strong interaction and covariance (Amundson et al., 2007). Biogeochemical studies demonstrate strong control of water and temperature on both elemental loss and chemical weathering rates across laboratory to watershed space and time-scales (Kump et al., 2000; Velbel, 1993; White et al., 1999). Similarly, soil climosequence studies demonstrate a strong correlation of climate to soil chemistry and mineral

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transformation in landscapes of low physical erosion (Chadwick et al., 2003; Dahlgren et al., 1997; Rasmussen et al., 2007; Williams et al., 2010). However, climosequence studies are generally designed to minimize the effects of physical erosion, and focus largely on the imprint of climate on soil properties as recorded by within-pedon mineral transformation and mass redistribution. In comparison, recent empirical geomorphic work at watershed and hillslope scales demonstrates that for moderately eroding landscapes, physical erosion exerts primary control on chemical weathering rates, with climate as a second order constraint (Hren et al., 2007; Jacobson et al., 2003; Riebe et al., 2004; West et al., 2005). This contrasts substantial watershed biogeochemistry work noting strong temperature correlation to weathering rates (Dorn and Brady, 1995; Turner et al., 2010; White and Blum, 1995), and recent geomorphic work suggesting strong climate relation to soil and saprolite chemical weathering (Dixon et al., 2009b). The complete nature of the climate–erosion interaction and its control on the biogeochemical processes of chemical mass loss, weathering rate, and regolith formation thus remains poorly defined.

At the hillslope scale, controls on the physical transport of regolith material have been explored using the concepts of transport- and weathering-limitation (Carson and Kirkby, 1972; Gilbert, 1877), with later work linking these regimes to large river basin denudation and solute chemistry (Stallard and Edmond, 1983). Similar notions have recently been used to describe and model the relation of physical erosion and mineral supply to chemical weathering rates, expressed simply as (Ferrier and Kirchner, 2008; Gabet and Mudd, 2009; Millot et al., 2002; West et al., 2005):  $W \propto E^\lambda$ , where  $W$  is chemical weathering rate,  $E$  is the physical erosion and assumed equivalent to the rate of mineral supply into the regolith under steady-state conditions, and  $\lambda$  is a dimensionless constant. Values of  $\lambda \approx 1$  are attributed to regimes where physical erosion limits chemical weathering, whereas  $\lambda < 1$  indicates regimes limited at least partially by chemical weathering kinetics. Others have suggested that relating chemical weathering to total landscape lowering or denudation ( $D$ , where  $D = E + W$ ) presents a better framework for identifying physical constraints on chemical weathering rates (Riebe et al., 2004). This may be particularly important for landscapes that experience substantial chemical weathering at depth in saprolite layers that are not physically transported (Dixon et al., 2009a). In these systems, the total chemical weathering rate ( $W_{total}$ ) is the sum of weathering occurring in upper soil layers susceptible to physical transport ( $W_{soil}$ ) and isovolumetric weathering occurring in saprolite ( $W_{sap}$ ) where:  $W_{total} = W_{soil} + W_{sap}$ , such that total landscape lowering or total denudation is equivalent to (Dixon et al., 2009a):  $D_{total} = E + W_{total}$ . The expression:  $W_{total} \propto D_{total}^\lambda$ , thus provides a proxy for the relative dominance of landscape lowering by physical and chemical processes. Here,  $\lambda$  carries much the same meaning as the previous discussion in that at  $\lambda \approx 1$ , the factor limiting denudation is physical erosion, whereas at  $\lambda < 1$  physical erosion dominates denudation and indicates that some factor, such as water availability, temperature, or solute composition kinetically limits chemical weathering (Maher, 2010; White and Brantley, 2003). Herein we follow this convention and use the terms erosion- and kinetic-limited to refer to these two end-members, with a transitional regime between the two limits.

Recent work modeling regolith production by coupling reactive transport with denudation suggests that the shape, form and degree of the regolith mineral depletion profile may be quantitatively related to the type of weathering regime (Lebedeva et al., 2007, 2010). This work represents a step towards bridging the shape and form of elemental depletion profiles with chemical weathering rates. However, extensive empirical data examining full regolith depletion profiles in the context of erosion- and kinetic limits and the climate–erosion–weathering interaction are lacking.

The objective of this study was to address this knowledge gap through synthesis of regolith and rock geochemistry, climate, and denudation data across a range of granitic terrains. We hypothesized

that water availability and temperature are the dominant controls on Na chemical depletion and weathering rates in locations of low to moderate physical erosion and minimal human disturbance. As a first order test of this hypothesis, we sought all available data for locations that could provide elemental concentrations from land surface down to and including bedrock for one lithology (granite) and one topographic condition (upland locations with low relief). With these data, we explored regolith transformation as a function of water availability, temperature and physical erosion with a specific focus on sodium as a biologically inert proxy for plagioclase weathering.

## 2. Materials and methods

### 2.1. Study sites and approach

This study was focused on linking regolith geochemical profiles and chemical weathering rates to climate and erosion data. Regolith is defined here to include soil and saprolite layers down to the depth of unweathered bedrock defined as the “depth of refusal”, the contact where regolith can no longer be excavated by hand, hydraulic core, or drill (Soil Survey Staff, 1999). The compiled data were limited to regolith developed on granitic rocks in locations little disturbed by human activities as reported for idealized 1-dimensional weathering profiles, i.e., upland, ridge-top positions with minimal to no colluvial material (Taylor and Eggleton, 2001) (Table A1). Ridge-top locations represent the simplest possible landscape positions with 1-dimensional vertical processes dominating soil production and chemical weathering (Brantley and White, 2009). Limiting data to one landscape position eliminates the effects of slope steepness, downslope transport, and colluvial layer thickness on regolith development (Yoo et al., 2007). Data were excluded only for cases where there were clear lithologic discontinuities in the regolith profile such as glacial till over bedrock, surface slope was greater than 7.5°, or where rock geochemistry was poorly constrained. The slope angle of 7.5° was chosen based on field and laboratory data indicating that slopes of less than 7.5° exhibit lower runoff coefficients and higher infiltration rates relative to steeper landscape positions that indicate water is more likely to infiltrate vertically and hence promote 1-dimensional regolith weathering (Chaplot and Le Bissonnais, 2000; Fox et al., 1997).

For the data included here, the sites were concentrated in the mid- to low-latitudes (18 to 40° N and S) with an average mean annual precipitation (MAP) that ranged from 370 to 4200 mm yr<sup>-1</sup> and average mean annual air temperature (MAT) that ranged from 3.9 to 30.0 °C (Table A1). Water availability was quantified using a simple dimensionless “humidity index”, equivalent to mean annual precipitation divided by mean annual potential evapotranspiration (PET): HI = MAP/PET, where HI is humidity index and both MAP and PET are in units of length per unit time. The humidity index, a standard approximation of annual water balance, is used extensively in soil science (Buol et al., 2003) and hydrology (Brutsaert, 2005). Humidity index values greater than one indicate positive annual water balance, whereas values less than one indicate negative annual water balance. Following hydrologic convention, we use the terms “energy-limited” when HI > 1 and precipitation exceeds the energy available for evapotranspiration, and “water-limited” when HI < 1 and evapotranspiration is limited by the amount of precipitation (Budyko, 1974). We recognize that an annual humidity index does not capture short-term variation of soil–water dynamics, water residence time or solute composition that are important mechanistic controls on mineral weathering over daily to annual time-scales (Maher, 2010). However, an annual humidity index provides a meaningful empirical proxy for decadal scale water availability that can provide insight as to the role of climate in regolith formation (Jenny, 1941).

The humidity index was calculated using site-specific precipitation data and potential evapotranspiration calculated from mean monthly temperature and latitude (Black, 1996; Thornthwaite, 1948). The Thornthwaite method requires only monthly mean temperature and

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