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# Lithological influences on contemporary and long-term regolith weathering at the Luquillo Critical Zone Observatory

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**Abstract**

Lithologic differences give rise to the differential weatherability of the Earth's surface and globally variable silicate weathering fluxes, which provide an important negative feedback on climate over geologic timescales. To isolate the influence of lithology on weathering rates and mechanisms, we compare two nearby catchments in the Luquillo Critical Zone Observatory in Puerto Rico, which have similar climate history, relief and vegetation, but differ in bedrock lithology. Regolith and pore water samples with depth were collected from two ridgetops and at three sites along a slope transect in the volcaniclastic Bisley catchment and compared to existing data from the granitic Río Icacos catchment. The depth variations of solid-state and pore water chemistry and quantitative mineralogy were used to calculate mass transfer (tau) and weathering solute profiles, which in turn were used to determine weathering mechanisms and to estimate weathering rates.

Regolith formed on both lithologies is highly leached of most labile elements, although Mg and K are less depleted in the granitic than in the volcaniclastic profiles, reflecting residual biotite in the granitic regolith not present in the volcaniclastics. Profiles of both lithologies that terminate at bedrock corestones are less weathered at depth, near the rock-regolith interfaces. Mg fluxes in the volcaniclastics derive primarily from dissolution of chlorite near the rock-regolith interface and from dissolution of illite and secondary phases in the upper regolith, whereas in the granitic profile, Mg and K fluxes derive from biotite dissolution. Long-term mineral dissolution rates and weathering fluxes were determined by integrating mass losses over the thickness of solid-state weathering fronts, and are therefore averages over the timescale of regolith development. Resulting long-term dissolution rates for minerals in the volcaniclastic regolith include chlorite: $8.9 \times 10^{-14}$ mol m$^{-2}$ s$^{-1}$, illite: $2.1 \times 10^{-14}$ mol m$^{-2}$ s$^{-1}$ and kaolinite: $4.0 \times 10^{-14}$ mol m$^{-2}$ s$^{-1}$. Long-term weathering fluxes are several orders of magnitude lower in the granitic regolith than in the volcaniclastic, despite higher abundances of several elements in the granitic regolith. Contemporary weathering fluxes were determined from net (rain-corrected) solute profiles and thus represent rates over the residence time of water in the regolith. Contemporary weathering fluxes within the granitic regolith are similar to the long-term fluxes. In contrast, the long-term fluxes are faster than the contemporary fluxes in the volcaniclastic regolith. Contemporary fluxes in the granitic regolith are generally also slightly faster than in the volcaniclastic. The differences in weathering fluxes over space and time between these two watersheds indicate significant lithologic control of chemical weathering mechanisms and rates.

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## 1. INTRODUCTION

Silicate mineral weathering in the critical zone is a primary source of solutes and sediments to streams and oceans where they contribute to the sequestration of $CO_2$, providing a negative climate feedback over geologic time (e.g., Berner and Berner, 1997). Therefore, the relative weatherability of Earth's surface (e.g., Kump et al., 2000) is a fundamental constraint on Earth's climate as well as on biogeochemical cycles. Relative weatherability and the lithologic mechanisms that produce silicate weathering products can only be identified and isolated by comparing weathering of different rock types while controlling for climate variability and other parameters (e.g., Berner, 2008). However, many previous studies of silicate weathering fluxes have not controlled for climate in this way (e.g., Bluth and Kump, 1994; White and Blum, 1995; Kump et al., 2000; Dessert et al., 2001).

Silicate weathering also provides mineral nutrients to soils and regolith and is an important factor in the rate of regolith formation and evolution. It is likely that the majority of exported solutes are liberated by weathering processes occurring deep in the critical zone (e.g., Rad et al., 2007; Kurtz et al., 2011; Schopka and Derry, 2012) where weathering-susceptible primary minerals are abundant and exposed to reactive fluids: at rock-regolith interfaces or within bedrock aquifers and fracture networks near the water table. Weathering at rock-regolith interfaces produces porosity and essential mineral nutrients (e.g., P, Mg, Ca, K, Fe), which support terrestrial ecosystems (Walker and Syers, 1976; Buss et al., 2005; Buss et al., 2008, 2010; Graham et al., 2010; Minyard et al., 2011; Hahm et al., 2014). Although near-surface and deep critical zone ecosystems may operate as largely independent nutrient cycles (Buss et al., 2005), especially in deep tropical regolith, they may still affect one another because (i) regolith formed at the rock-regolith interface will effectively move towards the surface as erosion lowers the ground surface over time (e.g., Brantley and White, 2009) and (ii) weathering reactions and soil organisms can alter the chemistry and reactivity of infiltrating water. For example, respiration of $O_2$ by soil microorganisms reduces the concentration of $O_2$ in pore water, which can slow down both geochemical and biological oxidation reactions (Fletcher et al., 2006; Buss et al., 2008; Bazilevskaya et al., 2013; Brantley et al., 2014).

In *in situ* critical zones, the link between the near-surface (classical soil) and deep zones (fractured and disaggregating bedrock) is saprolite. Here we use the term *saprolite* to refer to friable material weathered in place (not strictly limited to isovolumetrically weathered material), *soil* for the overlying, bioturbated, rooting zone and *regolith* as a general term encompassing soil, saprolite and any mobile, weathered, friable material (e.g., Taylor and Eggleton, 2001). Mineral weathering and biogeochemical processes within saprolite determine many characteristics of the overlying soil; indeed, saprolite has traditionally been considered parent material, or C-horizon, in classical soil science (Richter and Markewitz, 1995). Furthermore, as noted above, mineral weathering and biogeochemical processes within regolith also determine the reactivity of pore water that reaches the underlying bedrock, with implications for whole-rock weathering rates, regolith formation rates, watershed solute fluxes, and modulation of global $CO_2$. Mineral weathering reactions within regolith also produce nutrients and energy sources for resident microorganisms (e.g., Buss et al., 2005).

Despite the importance of silicate mineral weathering rates to myriad critical zone and global processes, relatively few field-based rates have been measured (see compilation in White and Buss, 2014) and most weathering studies of volcanic materials have focused on basaltic terrains. Different lithologies have different mineral assemblages and different characteristics (e.g., grain size) that may affect mineral weathering mechanisms and rates. Although andesite and basalt make up similar proportions of the global land surface, andesites are more common on tropical and subtropical islands and near active margins while many of the basaltic terrains worldwide are located on continents and at higher latitudes. Tropical watersheds, which cover only about 25% of the terrestrial surface, produce 65% of the dissolved silica flux to the oceans via mineral weathering processes (Stallard and Edmond, 1983; Meybeck, 1987). Volcanic islands, globally, contribute about 30% of the solute flux to the world's oceans despite representing only 9% of the terrestrial surface (Rad et al., 2007). Therefore, silicate weathering fluxes from low-latitude andesites may be more significant to global $CO_2$ consumption than fluxes from basalts (Goldsmith et al., 2010). However, measured watershed-scale silicate weathering fluxes in andesitic and basaltic watersheds are similar (McDowell and Asbury, 1994; Dessert et al., 2003 and references therein; Rad et al., 2006; Goldsmith et al., 2008, 2010).

The aforementioned andesitic weathering studies determined chemical weathering fluxes from concentration and discharge data for rivers draining catchments of interest, effectively providing watershed-averaged, endpoint views of chemical weathering from which weathering mechanisms and rates are inferred. That approach is complemented by studies that examine chemical weathering processes within weathering profiles, i.e., *in situ*, which provide mechanistic information enabling determination of mineral-specific reaction rates (Brantley and White, 2009; White and Buss, 2014) as well as providing spatially resolved information about mineral nutrient availability in terrestrial ecosystems (e.g., Buss et al., 2005). Several studies of both types have been done in the Luquillo Mountains of Puerto Rico, in what is now the Luquillo Critical Zone Observatory (LCZO) (McDowell and Asbury, 1994; Murphy et al., 1998; White et al., 1998; Schulz and White, 1999; Buss et al., 2008, 2010; Stallard, 2012; Buss et al., 2013), mostly within the granitic Río Icacos watershed. Here we present weathering profile data from the andesitic, metavolcaniclastic Bisley catchment as a comparison to the neighboring granitic Río Icacos catchment to investigate the influence of lithology on regolith weathering processes and rates, and on mineral nutrient availability. Solid-state elemental and mineral abundances with depth in the regolith were used to estimate mineral weathering rates and elemental fluxes over the timescale of regolith development

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