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Investigating the complex interface where bedrock transforms to regolith

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1.1. Introduction

Many chemical, physical, and biological processes combine to transform bedrock to regolith. These processes occur across an interface over which the chemistry, particle size, and biotic inhabitants of the regolith change as the rock equilibrates to surface conditions. The interface is not planar but rather has a roughness that varies with scale of observation. We have been investigating the distribution and geometry of this bedrock-saprolite interface in two Critical Zone Observatories (CZOs) that differ drastically in composition, climate, and erosional regime. The first site lies on Rose Hill shale in central Pennsylvania (Shale Hills CZO), and the second on Rio Blanco quartz diorite in the Rio Blanco/Icacos watershed in Puerto Rico (Luquillo CZO) (Figure 1). For both sites, the bedrock-saprolite interface is manifested in i) element-depth distributions, and ii) rock fragment size-depth distributions. These distributions define the bedrock-regolith interface and may be related to the longitudinal profiles of the watershed streams.

1.2. Results: Depth distributions of element concentrations

The thickness of the bedrock-regolith interface can be equated to the depth interval over which reactions occur as meteoric fluids charged with O_2 and CO_2 infiltrate bedrock. As minerals dissolve, element concentration-depth profiles evolve to form depletion profiles (BRANTLEY and WHITE, 2009). Depletion profiles show a decrease in a normalized concentration upward (Figure 1 (BUSS et al., 2008; JIN et al., 2010)). Concentrations are normalized using the mass transfer coefficient τ to correct for loss or gain of other elements (ANDERSON et al., 2002). Minerals that become completely depleted ($\tau = -1$) at the land surface are said to have *completely developed* depletion profiles while those still present at the surface have *incompletely developed* profiles (BRANTLEY and WHITE, 2009). For example, in Figure 1, the K (in clays) and Na (in feldspar) comprise incompletely developed profiles at Shale Hills whereas Ca

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(in ankerite) is completely developed. In contrast, in Puerto Rico, the primary minerals shown demonstrate completely developed profiles. In both cases, quartz is present in the regolith, comprising an incompletely developed profile (not shown).

The thickness of depletion is affected by the chemistry and rate of fluid flow. Both the PA and Puerto Rico profiles in Figure 1 developed on ridgetops where precipitation > evaporation. For such a case, dissolution of mineral *A* can be modeled as occurring due to one-dimensional downward fluid flow through the porous material. The distance along the flow path over which *A* reacts is the *reaction front*. The thickness of the reaction front, l_{eq} , is approximated as the distance between the depth where mineral *A* is completely dissolved, i.e. $\tau = -1$, and the depth where 1/e ($\approx 1/3$) of concentration of *A* is dissolved, i.e. $\tau \approx -0.37$. In a system where transport is by diffusion and advection, the thickness of the reaction front depends upon the Darcy velocity *v*, the solubility of the mineral, C_{eq} , and the product of the rate constant *k* and the specific mineral surface area, *s*: $l_{eq} = 2\phi D/(v(W-1))$, where $W = [1 + 4ks\phi D/(C_{eq}v^2)]^{1/2}$ (LICHTNER, 1996). For pure advection, l_{eq} equals vC_e/ks . Here, ϕ equals porosity and *D* equals the diffusion coefficient. Although these equations were derived for one-component, one-phase systems, the dependence of l_{eq} on parameters is similar for multicomponent, multiphase systems (LICHTNER, 1996).

According to these equations, l_{eq} depends upon the rate constant. Furthermore, as long as mineral A demonstrates an incompletely developed profile, the rate of advance of the reaction front -- the weathering advance rate, ω -- also depends on kinetics. However, at complete depletion, ω no longer

depends upon kinetics but only on solubility (see, for example, Lichtner, 1996): $\omega = \frac{v(C_{eq} - C_0)}{\phi_s^0 \overline{V_s^{-1}}}$. Here

 $\overline{V_s}$ equals the molar volume of *A*, ϕ_s^0 equals the fraction of mineral *A* in the starting rock, and C_0 equals the concentration of dissolved *A* before dissolution commences. Furthermore, the time required for mineral *A* to completely disappear at the land surface, τ_s , varies with the rate constant *k*:

 $\tau_s = \frac{\overline{V}_s^{-1} \phi_s^0 C_{eq}}{ks(C_{eq} - C_0)}.$ The one-component, one-phase model of Lichtner and other similar models in the

literature (ORTOLEVA et al., 1986) are useful for non-eroding systems but have been extended to include erosion at steady-state (LEBEDEVA et al., 2010).

The solubilities and reaction kinetics of most silicates increase with decreasing pH below neutral. However in the models described above, pH is generally not explicitly incorporated. The value of pH could be included implicitly by choosing *k* for a given pH, but *k* does not change as pH changes along the flow path. In weathering systems, pH varies due to production of organic acids, CO_2 , or protons. Because weathering reactions of most carbonates and silicates consume protons, the pH of infiltrating fluids generally increase, slowing rates of dissolution along flow paths. However, the release and oxidation of Fe²⁺ to Fe³⁺ followed by precipitation acidifies solutions. As oxidizing fluids progress through rock containing minerals such as pyrite, therefore, the pH drops and weathering can accelerate in pyrite-containing as opposed to pyrite-free zones (LICHTNER, 1996). Weathering advance rates thus vary with the abundance of soluble carbonate, silicate, and ferrous iron-containing minerals.

For example, in Shale Hills, the most abundant dissolving silicate is illite. Dissolution of illite begins in rock beneath augerable regolith. At this depth, the bedrock has fractured into blocky, angular fragments decimeters in dimension. This zone, as well as ~24 meters of underlying bedrock, were

sampled by drilling at the northern ridgetop. The core contains significant carbonate or pyrite only at depths < 20 m (carbonate shown in Figure 1, pyrite not shown).

In contrast, the most abundant mineral in the Luquillo quartz diorite, plagioclase, begins to dissolve beneath the saprolite in the cm-thick "rindlets" or onionskin-like shells of rock that form during spheroidal weathering (Figure 1). The rindlets surround spheroidal corestones that are meters in diameter (Figure 2) (BUSS et al., 2008). Neither carbonate nor pyrite has been observed to be significant in the quartz diorite sampled in outcrop. However, pyrite was observed in a drilled core nearby. The core, drilled on Route 191, intersected saprolite and one set of ~40 rindlets at ~645 masl, i.e. 6 m below the road surface. Beneath the rindlets, no more spheroidal weathering features were observed (~30m total drilling depth). The pyrite was observed in patches below 20 m depth (i.e. 630 masl).

1.3. Results: Depth distributions of rock fragment size

Based on such depletion profiles, the bedrock-regolith interface is at least as thick as 25 m in both the Rose Hill shale and Rio Blanco quartz diorite. Most studies of weathering profiles have focused on such element concentration-depth curves. As minerals dissolve, however, the size of the reacting fragments also change. In an eroding ridgetop (e.g., Figure 1), these fragments move upward over geological time, becoming smaller. The distribution of particle size with depth also documents the bedrock-regolith interface and the rates of weathering (FLETCHER and BRANTLEY, 2010). Fletcher and Brantley proposed a model (Figure 2) that describes how the sizes of corestones decrease upward due to dissolution of volcaniclastic bedrock in a first-order catchment within the Rio Mameyes watershed. This watershed lies adjacent to the Rio Blanco/Icacos catchment in the Luquillo CZO. The thickness of the reaction front, inferred from the elevation interval over which exhumed corestones change in size, equals ~100 m (Figure 2, 3).

The size of quartz diorite corestones in the Rio Icacos watershed can also be observed where erosion has exposed them on hillslopes (measurements from the Quebrada Guaba and El Toro trail shown in Figure 3). Corestones decrease in size at higher elevations (Figure 2). Like the Mameyes profile, the corestones disappear at the highest elevations so that only saprolite covers the ridgetops. We infer that the thickness of this depth interval of corestone size change is related to the balance between rates of weathering and erosion as well as the initial corestone size. This latter dimension is most likely dictated by fracture spacing in the underlying rock (Fletcher and Brantley, 2010).

In Shale Hills, rock fragments also decrease in size upward, although our data set currently is limited to the trend in the augerable regolith itself (Figure 2). These fragments form from the Rose Hill bedrock due to fracturing and water infiltration. Like Luquillo, we infer that fracture spacing of Rose Hill shale bedrock determines the fragment size at the base of regolith.

1.4. Discussion: Distribution of the weathering interface

These element concentration and size profiles are distributed within the watershed in patterns related to topography. For example, throughout most of the Shale Hills first-order catchment, we have observed that rock fragments of size larger than 1 cm are only rarely observed at the land surface, except in the presence of recent tree throws. Furthermore, the longitudinal profile along the Shale Hills channel is concave-upward and only in one of the highest points along this profile are rock fragments common at the land surface.

In contrast, as noted above, corestones are generally not observed at ridgetops in the Rio Icacos/Blanco watershed but are observed along hillsides. Furthermore, the longitudinal profile of the Rio Icacos/Blanco differs from that of the normal concave-upward profile of most monolithologic river incisions because it shows a break in slope (knickpoint) at ~600 masl (Figure 4). Above that elevation, the channel is lined by rindletted corestones in the walls and bed of the channel. At higher elevations the stream appears and disappears as it moves in and out of the partially buried corestone matrix. The channel slope is comparatively gentle with sand deposits sometimes burying corestones. Rindletted corestones are observed throughout the channel from ~650 to 600 masl while the lower Icacos below 465 m is bedrock-lined. No observations have been made on the Icacos between 600 and 465 masl due to difficult terrain. However, the longitudinal profile of one of Rio Blanco's other tributaries (Rio Sabana) exhibits the same break in slope. In that river, the channel below 600 m is mostly bedrock-lined. The break in slope throughout the Rio Blanco watershed at ~600 m is tentatively attributed to the change from incision into bedrock to incision into rindletted corestone-bedrock.

One inference from this working hypothesis might be that spheroidal weathering is faster at elevations > 600 m. Corroborating this, the ~ 30 m deep core ($\sim 645-620$ masl) drilled into the quartz diorite revealed only one set of rindlets at 5-5.8 m depth (645 masl). The fast weathering results in extensive deposition of sand above ~ 620 masl, perhaps consistent with transport-limitation above that elevation. Presumably, the fast rates of weathering at higher elevations are due to higher rainfall and higher porefluid velocities, as described by equations introduced from Lichtner (1996) above.

1.4. Conclusions

Both element-depth and fragment size-depth curves document the interface where bedrock transforms to regolith, and this interface is more than 20 meters in thickness in both Pennsylvania and Puerto Rico. Such weathering thicknesses, modeled as a function of one-dimensional fluid flow, are dictated by the balance between rates of weathering and erosion. For any given set of conditions, profiles may evolve toward a steady state where the advance rates of reaction fronts are coupled to one another by conditions on porefluid chemistry and particle size (FLETCHER and BRANTLEY, 2010). Given the importance of pH as a master variable and the importance of biota in affecting pH, this coupling may be catalyzed by biotic activity at the surface and at depth.

The one-dimensional flow models summarized here are consistent with weathering advance rates that vary with equilibrium solubility and porefluid velocities (and not reaction kinetics). However, fluid flow is, of course, not strictly downward beneath ridgetops. Permeability of regolith changes as particle size and bulk density changes with depth. Thus, both downward and lateral flow occurs. In fact, lateral flow is probably often accelerated at reaction fronts because reactions change permeability (Figure 3, right panel). Where lateral flow removes a significant fraction of the fluid from the downward flow, the weathering advance rate in underlying zones decreases compared to the theoretical one-dimensional system. Therefore, the rate of weathering advance is affected by the three-dimensional distribution of reaction zones that affect permeability across the watershed. We need to develop quantitative models of such phenomena over a range of spatial and temporal scales.

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Figure captions

Figure 1. Reaction fronts for mineral reactions at ridgetops in the Shale Hills Critical Zone Observatory (left, on Rose Hill shale) and the Luquillo Critical Zone Observatory (right, on Rio Blanco quartz diorite). Reaction fronts are plotted as τ_j values where *j* is an element (left) or a mineral (right). This value varies from 0 to -1 for 0 to 100% depletion (ANDERSON et al., 2002). In Luquillo, spheroidal weathering forms onionskin-like rindlets as plagioclase and other minerals dissolve (BUSS et al., 2008).

Figure 2. (Left) Three model simulations of how corestones become detached at the bedrock-regolith interface and diminish in size upward toward the land surface (top horizontal boundary) as they transform to saprolite. In the model, corestones become smaller as albite dissolves to form kaolinite (y axis = dimensionless depth, x axis = dimensionless size of corestone). The bedrock fracture spacing determines the initial corestone size. Model developed by Ray Fletcher (FLETCHER and BRANTLEY, 2010). (Middle) Corestone size plotted versus elevation for the Rio Icacos watershed. (Right) Rock fragment size plotted versus depth for a ridgetop soil at Shale Hills.

Figure 3. When the bedrock-regolith interface created by scenarios such as shown in Figure 2 is eroded by channel incision, hillslopes may reveal the distribution of subsurface weathering corestones (left panel). The model shown in Figure 2 incorporates only one-dimensional downward flow. Actually, downward infiltrating water may cause flow in subsurface that is lateral as shown on right panel.

Figure 4. Longitudinal profile of the Rio Icacos/Blanco. Length scale across the figure is 25 km (PIKE et al., 2010).