

Physical and Chemical Controls on the Critical Zone



Road cut in Precambrian Boulder Creek granodiorite, showing weathered rock under a thin regolith

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Geochemists have long recognized a correlation between rates of physical denudation and chemical weathering. What underlies this correlation? The Critical Zone can be considered as a feed-through reactor. Downward advance of the weathering front brings unweathered rock into the reactor. Fluids are supplied through precipitation. The reactor is stirred at the top by biological and physical processes. The balance between advance of the weathering front by mechanical and chemical processes and mass loss by denudation fixes the thickness of the Critical Zone reactor. The internal structure of this reactor is controlled by physical processes that create surface area, determine flow paths, and set the residence time of material in the Critical Zone. All of these impact chemical weathering flux.

KEYWORDS: Critical Zone, weathering, erosion, soil, regolith, denudation

INTRODUCTION

The breakdown of rock at the Earth's surface produces a thin, porous covering, recently labeled the Critical Zone (CZ) (NRC 2001). Within it, rocks, soil, atmospheric gases, and meteoric water interact. This geologic-atmospheric-hydrologic interface provides the primary habitat for terrestrial life. The CZ displays tremendous heterogeneity both vertically, manifested in distinguishable layers of weathered rock, regolith, and soil, and laterally, by the diversity of landscapes and the distribution of soils across them. These variations reflect chemical and mechanical processes that drive the weathering front into unaltered rock, as well as physical erosion and chemical denudation processes that remove mass from the land surface and the interior of the CZ. Chemical weathering is driven by thermodynamic disequilibrium relative to conditions under which minerals form. Mechanical weathering and physical erosion are driven by stress gradients that break or move material. These stress gradients may arise from gravitational stress, phase transformations (such as ice growth, salt precipitation, oxidation or hydration of minerals), or biologic agents (roots, burrowing animals, worms, and the like). How do these chemical and mechanical processes interact to produce the CZ and its lateral variations?

Knowledge gained from studying the link between mechanical and chemical weathering contributes to understanding how weathering and erosion affect atmospheric CO₂ and climate. Early climate models explored a negative feedback

in which chemical weathering rates depend on temperature and temperature depends on CO₂ (Walker et al. 1981). A later alternative proposed that uplift and erosion control chemical weathering and hence CO₂ and climate (Raymo and Ruddiman 1992). Recent work emphasizes rock type, in addition to temperature and erosion, in controlling chemical weathering rates and climate (Dupré et al. 2003). These global models rely on largely empirical correlations among chemical weathering, climate, and physical process rates. Rarely considered in these assessments of chemical weathering flux—the chemical weathering rate per unit

land surface area—are the influences of variations in development of the CZ and the reactive surface area within it.

The balance between mechanical and chemical contributions to weathering determines how the CZ forms. Geomorphologists often consider weathering to be preparation of material for transport, primarily by mechanical breakdown into transportable blocks or possibly by chemical weakening. Transportable material is “regolith.” Physical erosion rates are either “transport limited,” where regolith is deep, or “weathering limited,” where they are controlled by rates of regolith production (Carson and Kirkby 1972). There are two geomorphic models for the rate of regolith production: (1) rate decreases exponentially with overlying regolith thickness, and (2) rate is at its maximum under some thickness of regolith, the so-called “humped” function (Heimsath et al. 1999). Evidence for the humped function comes from observation of the highest rates for chemical and physical processes (frost cracking, biotic disturbance) below the surface (Gilbert 1877; Anderson 1998; Gabet et al. 2003). Wilkinson et al. (2005) suggest that a “shifting mosaic” of vegetation may produce the appearance of exponential dependence of regolith production on thickness. In these models, regolith thickness stands as an indicator for the various physical and chemical mechanisms that break down rock.

The concepts of weathering-limited and transport-limited erosion were embraced by Stallard and Edmond (1983) in their analysis of landscape control of chemical weathering fluxes in the Amazon. They recognized that weathering-limited environments, which have little or no soil, deliver relatively unaltered sediment to streams. These landscapes produce high chemical-weathering fluxes, presumably due to highly weatherable minerals at the surface. Soil-mantled, transport-limited environments are more likely to yield highly weathered sediments and produce low solute fluxes.

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Geochemical controls on global weathering fluxes and physical controls on erosion are interrelated. Both geochemical and physical processes decay rock, each process feeding back on the other. It is time to move beyond correlations of weathering and erosion or regolith thickness and understand how these processes interact to shape the CZ and fluxes from it.

THE CRITICAL ZONE: A FEED-THROUGH REACTOR

One can think of the CZ as a feed-through reactor: in a region of net denudation or of topographic steady state (uplift balanced by erosion), solid material enters the reactor through its lower boundary, while solid sediment products are skimmed off the top by erosion and dissolved products leak out through its porous sides (Fig. 1). This conceptual model differs from a net depositional system, in which fresh material is added at the surface. A parcel of rock can be thought to move vertically through this system, fracturing and undergoing chemical weathering, until it reaches the rock–regolith boundary. Freed from surrounding rock, the parcel then moves stochastically, as it is mechanically and biomechanically impelled through the soil and moved downslope (Mudd and Furbish 2006; Kaste et al. 2007), undergoing abiotic weathering and biologically driven nutrient extraction (Burford et al. 2003). Some components may be retained within the reactor by uptake and cycling between vegetation and organic matter (e.g. Moulton et al. 2000).

Solutions produced from percolating rainwater, renewed with each rainstorm, bathe the CZ reactor. These are enriched with organic exudates and charged with dissolved gases (notably CO₂ and O₂). Water percolates at rates controlled by the precipitation (or infiltration) rate and by the hydraulic conductivity of the soil. Such conductivity can be highly variable depending on the material, its history, and the hydrologic conditions. Mineral dissolution or rock fracturing can increase permeability, while clay formation can reduce it. Unsaturated hydraulic conductivity varies by orders of magnitude with slight changes in water content. Mechanical and biomechanical processes open new avenues for water flow through rock, hence exposing new surfaces to reactive weathering solutions, moving the weathering front down, and bringing new material into the reactor. Thus, the vertical extent of the CZ—the size of the reactor—reflects the relative rates of downward weathering-front advance and erosion from the top.

Where in the Critical Zone Does Chemical Weathering Occur?

From a theoretical perspective, the composition, age, and surface area of minerals, as well as the temperature, flux, and composition of solutions, are important controls on intrinsic chemical weathering rates. These parameters vary with depth. Gradients through the CZ in solution parameters such as pH, dissolved Al, and H₂CO₃ influence reaction kinetics, while gradients in hydraulic conductivity, controlled by porosity, pore size, and water content, affect transport. A CZ reactor does not generally provide optimal chemical-weathering conditions. The fluid is likely most reactive and the matrix most permeable near the top of the reactor, while the minerals most prone to weathering are usually at the bottom of the reactor. How do these competing effects play out?

Differing system behaviors are reported in the literature. In the warm, wet, steep terrain of Luquillo Experimental Forest, Puerto Rico, complete weathering of plagioclase occurs at the bedrock–saprolite interface, while more resistant minerals survive farther up into the CZ (White et al. 1998). The silicate weathering flux in Luquillo is the highest recorded. In contrast, in the equally warm and wet Nsimi watershed, south Cameroon, silicate weathering fluxes are very low (Braun et al. 2005). The difference between these granitoid systems appears to be hydrologic. In Luquillo, the water table is below the saprolite, and water flushes rapidly through the profile. In Nsimi, high rainfall and low relief keep the water table above the saprolite, and water flow at the weathering front is sluggish. Another warm, wet, granitoid system occurs in Sri Lanka. Despite steep terrain, chemical weathering fluxes are low (von Blanckenburg et al. 2004), comparable to the swampy Nsimi catchment. In Sri Lanka, weathering appears to be hampered by low physical erosion rates, a consequence of tectonic quiescence.

Access of rock to solutions seems to control weathering fluxes from a headwater catchment near Coos Bay, Oregon Coast Range, USA. There, half of the solute flux is derived from fractured rock, despite a highly porous but hydrologically unsaturated regolith rich in primary minerals (Anderson and Dietrich 2001). Dissolved silica fluxes at Coos Bay are

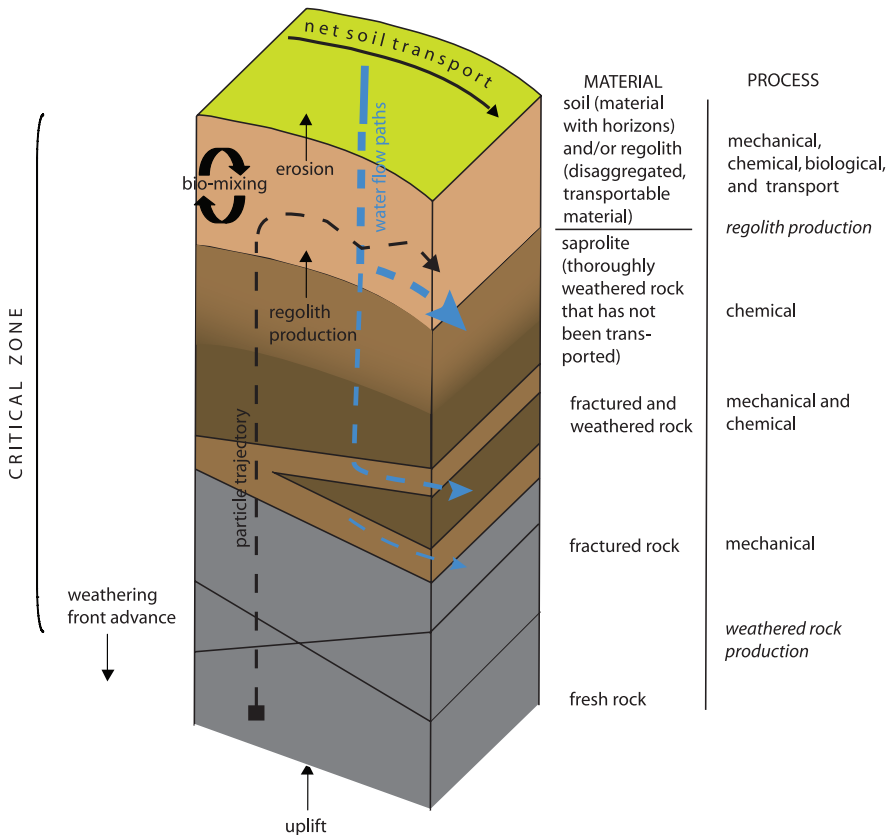


FIGURE 1 Vertical layering within the Critical Zone, showing rock parcel trajectory (black dashed line) and water flow path (blue dashed line) in a system with minimal addition of solid material at the top. Not all layers are present in all systems. Chemical weathering flux varies with depth and is controlled by mineral and weathering solution reactivity and water flow rate. In depositional systems, such as loess, volcanic ash falls, floodplains, and footslopes, particle trajectory is downwards relative to the surface and chemical flux can be highest at the top of the reactor where minerals are freshest.

nearly as high as in Luquillo (Anderson et al. 2002), despite lower temperatures and rainfall, illustrating the importance of access of aggressive water to unweathered minerals in controlling weathering fluxes.

Residence Time for Rock in the Reactor

The idea that the supply of fresh rock to the weathering environment controls chemical weathering flux has been around for more than a century. In a steady-state landscape, where rock uplift is balanced by denudation and the thickness of the weathered profile is steady, the rate of material supply controls the residence time of a rock parcel in the CZ. Under these commonly assumed conditions, the residence time is given by $t = Z/\dot{u}$ where Z represents CZ thickness and \dot{u} the rock uplift rate (or equivalently, total denudation rate). Waldbauer and Chamberlain (2005) termed this the effective surface age, a name that applies equally well at the mineral-grain scale as at the landscape scale. Note that this residence time is conveniently determined from cosmogenic nuclide concentrations at the top of a steadily eroding profile.

Several have argued that a shorter residence time for rock in the CZ, driven by faster erosion rates, produces higher chemical weathering solute yields (Anderson et al. 2002; Riebe et al. 2004; Waldbauer and Chamberlain 2005). The reasoning, similar to that of Stallard and Edmond (1983), is that where rock moves rapidly through the CZ, more weatherable minerals are accessible, mechanical breakdown of rock is faster, and mineral weathering rates are higher (White and Brantley 2003). Waldbauer and Chamberlain (2005) developed a model of chemical weathering flux as a function of rock uplift rate, illustrating direct correlation with mineral weatherability.

Setting the Pace of Chemical Denudation

What controls whether a mineral will weather in the CZ or survive for physical erosion? Mineral dissolution rates exhibit a dynamic range of about six orders of magnitude from the highest experimental to the lowest natural rate (FIG. 2). While some of this range is attributed to rougher mineral surfaces over time, the bulk of the decrease must be related to other time-dependent factors: (1) decrease in reactive surface area following diminished compositional and structural heterogeneities; (2) physical occlusion of surfaces by secondary precipitates and leached layers; and (3) extrinsic properties, such as the nature of the solution or its saturation state. For dilute conditions typical in experiments, reaction rate is limited by the mineral surface area and is independent of fluid composition and water flow rate. As solute concentrations approach equilibrium, reaction rate becomes limited by the rate of solute transport. This transport-limited chemical weathering happens in soil columns, where reaction rate increases with water percolation rate. Therefore, weathering under close-to-equilibrium conditions (the case in many field settings) is transport controlled (White and Brantley 2003). Water flow rate tends to decrease with depth in the CZ; this and progressive accumulation of weathering products would decrease chemical weathering rates at depth, but erosion of weathered material would tend to dampen this decrease.

One approach to determining the chemical denudation rate is to measure the total rate in the catchment using cosmogenic nuclides with the chemical depletion fraction, CDF (Riebe et al. 2004). CDF is the fraction of the total denudation resulting from chemical weathering, easily determined in a 1-D system from immobile element concentrations (such as zirconium and titanium) in the rock. A global survey using this approach in granitic catchments indicates a

correlation between physical erosion and chemical weathering rates (FIG. 3). Surveys based on riverine fluxes show a similar relationship (Gaillardet et al. 1999; Dupré et al. 2003; West et al. 2005) over a range of geological settings. Such compilations also show a relationship between chemical weathering flux and runoff, and to a lesser extent, temperature (e.g. Oliva et al. 2003). Silicate weathering flux is responsible for ~10% of the total (physical + chemical) denudation rate. This confirms that silicate weathering flux is affected by physical denudation rate, but depends only weakly on climate. The link to climate is mainly through precipitation-driven groundwater flow.

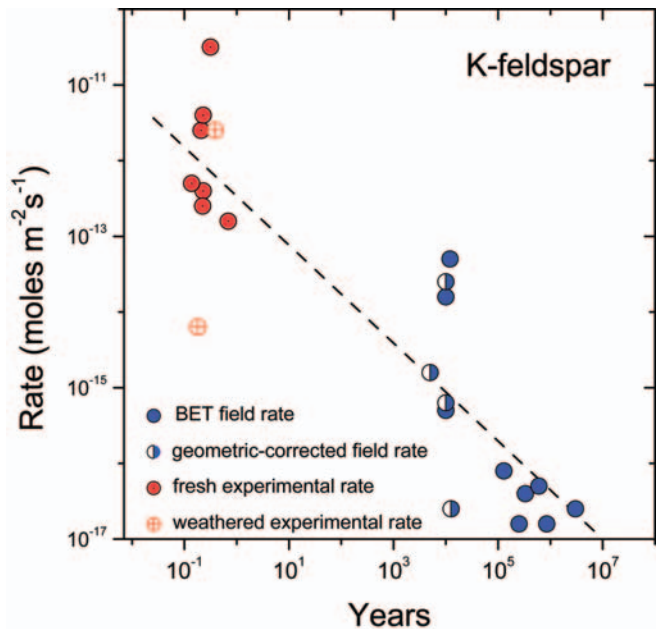


FIGURE 2 K-feldspar weathering rate as a function of time. Orders-of-magnitude lower field-based rates suggest external controls (e.g. pore-fluid saturation), which are in turn determined by physical hillslope processes (AFTER WHITE AND BRANTLEY 2003; REPRODUCED WITH PERMISSION FROM ELSEVIER).

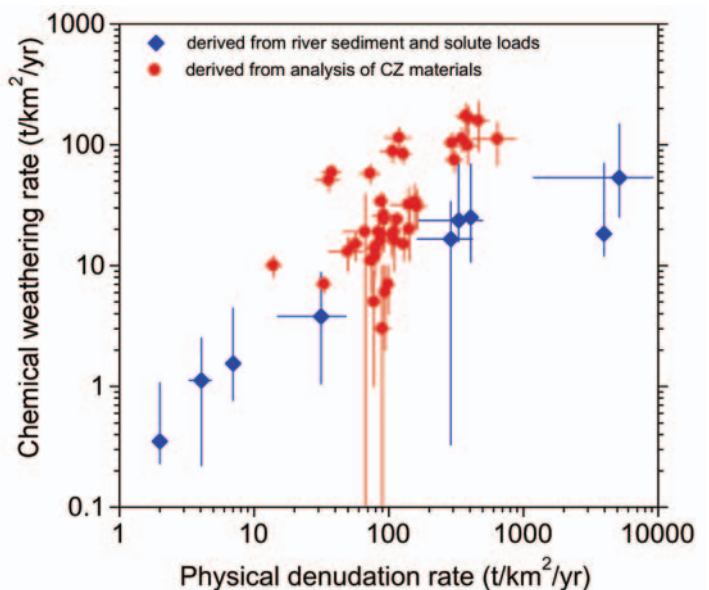


FIGURE 3 Comparison of flux rates determined from material in rivers (Millot et al. 2002) and from the residual CZ material (Riebe et al. 2004). Flux in the CZ was determined from solid-phase analysis using immobile elements and cosmogenic nuclides. Error bars for both data sets were taken from the published data sources.

Advancing the Weathering Front

Perhaps the most crucial physical-chemical interactions occur at the base of the CZ, where the weathering front advances into fresh rock. The difficulty in accessing this interface hampers understanding it. Chemical weathering control is illustrated in granite bedrock at Panola, USA (White 2005). Alteration of plagioclase to kaolinite can be seen in weathered rock 7.5 m below the surface, while adjacent orthoclase feldspar and biotite are untouched (Fig. 4). Such a pattern is attributed to small differences in mineral solubilities, despite similar kinetic reaction rates at far from saturation conditions. Percolating soil water (water in the soil above the groundwater table) reaches saturation with respect to K-feldspar but remains unsaturated with respect to plagioclase. Low permeability in the weathered rock limits water access; the saturation state of water at the base of the CZ controls the advance of the weathering front.

Fletcher et al. (2006) explored complex coupling between chemical and mechanical processes that advance the weathering front through spheroidal weathering of granite. Increased volume from oxidation of iron in biotite exerts stresses, ultimately fracturing the rock, forming multiple, onion-skin layers separated by cracks. Although diffusion of oxygen controls initial chemical weathering, the flow paths created by fractures allow rapid transport of oxygen and water deeper into the unweathered core. The rate of advance

of the weathering front is linear with time, rather than a function of the square root of time, as expected for true diffusion. Fluids are supplied and solutes removed through artery-like fractures. Thus, the slow diffusion of water through the rock matrix can operate over the short distances between fractures rather than over the whole thickness of the saprolite.

Fractures are undoubtedly important in advancing the weathering front. Tectonic processes likely control fracture density below the CZ (Molnar et al. 2007), and high fracture density predisposes rock for weathering and erosion. Much of the emphasis in discussions of weathering and climate has been on the effects of mountain building and erosion. Hovius and von Blanckenburg (2007) speculate that it is the rate and geometry of active tectonic faulting that accelerate physical, and therefore also chemical, denudation, whether faulting takes place in mountain belts or in extensional settings. Faulting potentially initiates two processes that advance the weathering front. First, faulting can induce rock fracturing, and second, faulting can trigger landscape readjustment by changing river base level, enhancing erosion.

Hovius and von Blanckenburg (2007) suggest two chains of consequences resulting from faulting:

- ① *Mechanical chain:* Faulting → fall in relative base level → incision of the drainage network → enhancement of soil erosion and rates of physical denudation and sediment production
- ② *Chemical chain:* Faulting → fall in relative base level → incision of the drainage network → development of new groundwater flow paths → stronger dilution and less saturation of soil pore water → enhanced rates of mineral dissolution → higher rates of chemical weathering and soil production

The influence of tectonic control on silicate weathering is illustrated in Sri Lanka, where very low rates of weathering and erosion have been observed, counter to expectations of behavior under conditions of high relief, hot climate, and high precipitation (von Blanckenburg et al. 2004). Sri Lanka, and the similar case of the Guyana Shield (Edmond et al. 1995), must be seen in light of the absence of tectonic forcing. Thick, clay-rich, weathered layers shield the bedrock from corrosive fluids. The absence of recent tectonic forcing, also characteristic of cratons, prevents high rates of weathering and denudation.

The Critical Zone on a Slope

We now consider the third dimension, that is, lateral transfer of material through the CZ. Lateral transfer affects the disarticulated and disaggregated material found in regolith and soil, not the deeper parts of the CZ. The importance of slope processes is recognized in the study of soil catenas, the distribution of soils on slopes. Transport processes collectively called *creep* move soil down slope, usually producing a velocity profile (averaged over many displacements associated with biotic or mechanical disturbances) that is fastest at the surface and declines with depth. One manifestation of creep is its impact on the chemical depletion factor (CDF). Yoo et al. (2007) show that this extra source of weathered material (enriched in immobile elements) must be factored into CDF calculations; otherwise, the CDF can be significantly overestimated.

Several models now exist that track regolith production, sediment transport, and chemical weathering on a hillslope (Mudd and Furbish 2006; Yoo et al. 2007). In steady state, such models predict that the mean age of the soil column is uniform everywhere on the hillslope, so chemical evolution and soil age should not vary with slope position. These models are not yet sophisticated enough to capture the differences in hydrologic conditions down a slope, thought to

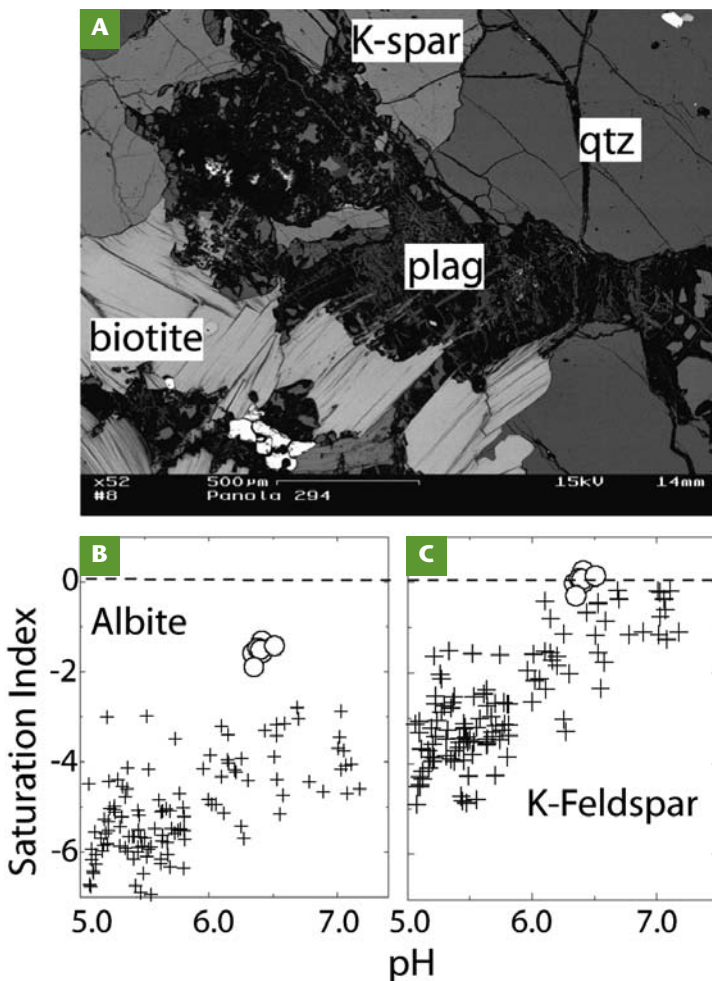


FIGURE 4 (A) Scanning electron microscope image of weathered granite, sampled 7.5 m below the surface at Panola, Georgia, USA. It shows altered plagioclase with pristine K-feldspar and biotite. (B and C) Soil water (crosses) and groundwater (circles) are far from saturation (dotted line) for albite, but groundwater is saturated with respect to K-feldspar (FROM WHITE 2005).

be important in soil catenas, and are not field tested. Clearly much remains to be explored with respect to sediment movement and weathering processes.

CONCLUDING THOUGHTS

The CZ behaves as a feed-through reactor, in which physical and chemical processes, often associated with biological agents, work together to digest and move material. The thickness of the reactor, the character of the weathered sediment and the solute “products,” and the rate at which they are formed depend on the rate of multiple chemical and physical processes within the reactor. Processes operating from tectonic (faulting, joint formation below the CZ) through hillslope (creep) to mineral scales (phase transformations inducing stresses that promote microcrack growth) all play a role in defining the chemical weathering rate. These processes control residence time of material in the CZ, the

rate of water flow, the saturation state of water, and the size of rock particles in the regolith, all of which influence the chemical weathering flux. Unraveling the links operating over a range of scales will require a shift in research approach from one of correlating observations to one of understanding the mechanical and chemical processes.

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