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Mineralogical transformations set slow weathering rates in low-porosity metamorphic bedrock on mountain slopes in a tropical climate

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ABSTRACT

In the Sri Lankan Highlands erosion and chemical weathering rates are among the lowest for global mountain denudation. In this tropical humid setting, highly weathered deep saprolite profiles have developed from high-grade metamorphic charnockite during spheroidal weathering of the bedrock. The spheroidal weathering produces rounded corestones and spalled rindlets at the rock–saprolite interface. We used detailed textural, mineralogical and chemical analyses to reconstruct the sequence of weathering reactions and their causes. The first mineral attacked by weathering was found to be pyroxene initiated by *in situ* Fe oxidation. Volumetric calculations suggest that this oxidation leads to the generation of porosity due to the formation of micro-fractures allowing for fluid transport and subsequent dissolution of biotite and plagioclase. The rapid ensuing plagioclase weathering leads to formation of high secondary porosity in the corestone over a distance of only a few cm and eventually to the final disaggregation of bedrock to saprolite. The first secondary phases are oxides or amorphous precipitates from which secondary minerals (mainly gibbsite, kaolinite and goethite) form. As oxidation is the first weathering reaction, the supply of O_2 is a rate-limiting factor for chemical weathering. Hence, the supply of O_2 and its consumption at depth connects processes at the weathering front with those at the Earth's surface in a feedback mechanism. The strength of the feedback depends on the relative weight of advective *versus* diffusive transport of O_2 through the weathering profile. The feedback will be stronger with dominating diffusive transport. The low weathering rate is explained by the nature of this feedback that is ultimately dependent on the transport of O_2 through the whole regolith, and on lithological factors such as low bedrock porosity and the amount of Fe-bearing primary minerals. Tectonic quiescence in this region and low pre-development erosion rate (attributed to a dense vegetation cover) minimize the rejuvenation of the thick and cohesive regolith column, finally leading to low denudation rates.

1. Introduction

The fact that most of the continental areas are covered by regolith (composed of the mobile soil layer overlying the *in situ* weathered saprolite) suggests that over millennial time scales removal of regolith material by erosion is mostly balanced by its production through bedrock weathering. This observation hints at the existence of a feedback between regolith depth and rates of weathering and erosion processes that produce and destroy regolith, respectively (Carson and Kirkby, 1972; Heimsath et al., 1997). The existence of this feedback is also suggested by the empirical negative relationship between soil production and soil thickness, the so-called “soil production function” (*e.g.*, Heimsath et al., 1997, 2009). The “soil production function” has been established for the mobile soil layer. Models for regolith development have suggested that this relationship might extend through the entire regolith (Lebedeva et al., 2010). The fact that a negative feedback between regolith thickness and weathering rate exists is an implicit conclusion of the observation that weathering rates are generally low where thick regolith prevails — a phenomenon commonly lumped into the term “soil shielding” (Goddéris et al., 2008; Hartmann et al., 2014; Stallard, 1995). However, alternative views suggest that in tectonically quiescent cratons, this feedback is absent such that regolith production outpaces erosion, leading to continuously thickening weathering profiles (Lebedeva et al., 2010).

In order to develop an improved understanding of the mechanisms leading to these relationships, it is critical (i) to examine in detail the

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mineralogical transformations occurring at the “weathering front”, where rock is first converted into saprolite; (ii) to identify the reactive phases (O_2, protons, acids, complexing agents) that limit the rate of these mineralogical transformations; and (iii) to characterize the formation of pathways, such as fractures and macro-cracks or connected pore spaces that support transport of reactive phases to the weathering front. In particular, an oxidative process for spheroidal fracturing was proposed by Buss et al. (2008), Fletcher et al. (2006), and Lebedeva et al. (2007), where Fe(II) in bedrock minerals is oxidized after O_2 transport from the soil surface into the rock. Oxidative weathering produces a specific volume increase of the solid material and thus builds up strain that fractures the rock. Similarly, precipitation of secondary clay minerals following dissolution of primary minerals was also proposed to result in strain build-up and rock fracturing (Jamtveit et al., 2008, 2011; Røyne et al., 2008; Rudge et al., 2010). In contrast to pathways that require the generation of strain, the access of reactive phases from the surface to the weathering front might also be promoted by the formation of secondary porosity by primary mineral dissolution and precipitation of secondary minerals with a lower molar volume (Lebedeva et al., 2007; Navarre-Sitchler et al., 2011; Sak et al., 2010; Velbel, 1993). All these models imply that the supply of a reactive compound from the atmosphere (*e.g.*, O_2) or from the upper regolith layers (*e.g.*, organic acids) is necessary to the downward propagation of the weathering front, thereby providing a negative feedback between the regolith thickness and mineral dissolution taking place at depth (Fletcher et al., 2006). Alternative models suggest that the lowering of the water table induced by channel incision promotes the downward propagation of the weathering front (Edmond et al., 1995; Rempe and Dietrich, 2014). Such a mechanism also requires the formation of fractures and open pore spaces for corrosive fluids to reach unweathered minerals in low-porosity bedrock. Evidence is still lacking about the processes that generate these fluid pathways and about their relative contribution to the weathering front advance.

Exploring the way these pathways are generated is important because their understanding is fundamental to quantitative models of the weathering front advance (Bazilevskaya et al., 2013, 2015; Brantley et al., 2008; Fletcher et al., 2006; Goddéris et al., 2006; Lebedeva et al., 2007, 2010; Moore et al., 2012; Navarre-Sitchler et al., 2011). These models essentially characterize the transport of solutes and gases through regolith profiles and the participating weathering reactions. The successful application of these models relies on an accurate knowledge of the weathering system's transport properties (*e.g.*, soil and saprolite permeability) and chemical parameters (*e.g.*, kinetics of dissolution and precipitation, or equilibrium constants). The nature and properties of the secondary precipitates forming during weathering (*i.e.*, their mineralogy, crystallinity, or ability to form aggregates) exert a strong control on the outputs of such models (Maher et al., 2009). In particular, many weathering reactions have been reported to produce amorphous intermediate phases rather than forming crystalline clays directly from primary minerals (Chadwick and Chorover, 2001; Dahlgren et al., 1997; Hellmann et al., 2012; Steefel and van Cappellen, 1990). Hence, the relevance of these weathering model predictions needs to be evaluated against thorough field observations across a range of different lithological and geomorphological contexts.

A weathering profile at Hakgala in the Central Highlands of Sri Lanka offers the opportunity to explore in detail the sequence of weathering processes from coherent bedrock to loose soil in a tropical mountain environment that is not currently subject to significant tectonic activity. Weathering and erosion rates, and the degree of chemical weathering have been quantified in great detail (Gunnell and Louchet, 2000; Hewawasam et al., 2003, 2013; Vanacker et al., 2007b; von Blanckenburg et al., 2004), providing boundary conditions on the relevant mass fluxes across the weathering system. Hewawasam et al. (2013) showed that in this setting weathering along hill slopes is slow, and operates at a steady state, *i.e.*, regolith removal by erosion balances regolith production by downward propagation of the weathering front. Such steady state is indicated by finding roughly similar weathering rates over two different time scales. The rates calculated from river dissolved loads (time scale *ca.* 10 years) agree with those from cosmogenic nuclides that are combined with chemical weathering indices in the profile (time scale *ca.* 10^4 years). Because the Hakgala weathering profile is a well-characterized, thick regolith profile representative of slow denudation-tropical settings that prevail on a large portion of the continental surface (Braun et al., 2009, 2012; Dequincey et al., 1999; Edmond et al., 1995; Hewawasam et al., 2013), it provides a perfect field site to study in detail the links between regolith properties and weathering at depth. In particular, the Hakgala weathering profile features corestones consisting of bedrock fragments remaining in the regolith that are progressively spheroidally weathered. As such, corestones include a core of bedrock and several layers around this core with increasing weathering intensities outward. They serve as small natural laboratories on which incipient weathering can be studied in detail (Buss et al., 2008; Ma et al., 2012; Sak et al., 2004, 2010). In fact, as the fronts of the respective weathering reactions move inwards with time, the spatial alignment also represents the state of weathering at different times, with outer layers having been exposed to weathering for longer times than inner layers.

In the present study, we build upon chemical and mineralogical analyses conducted by Hewawasam et al. (2013) who derived chemical mass fluxes, weathering degree, and the weathering front advance rate. Here, we further (i) identify the chemical reaction mechanisms and show the significance of amorphous secondary phases for the conversion of bedrock to loose material; (ii) identify textural changes between the compartments of the weathering profile; and (iii) discuss reactions that involve element fluxes and volume changes driving chemical weathering and the nature of the feedbacks between regolith thickness and propagation of the weathering front. With these new results we address the following questions: What are the mechanisms responsible for the slow propagation of the weathering front in deep regoliths under tropical climate? How is the propagation rate of the weathering front influenced by regolith transport properties? Why are low weathering rates in tropical mountain regions often associated with a thick regolith cover?

2. Study site and analytical methods

2.1. Study site and sampling

The Hakgala weathering profile (06.92923° N, 80.81834° E) is located on a fresh road cut on the road from Nuwara Elia to Welimada in the Central Highlands of Sri Lanka (please see Fig. 1, p. 204 in Hewawasam et al., 2013). The site is exposed to two monsoon seasons, mean annual precipitation is 5000 mm, and mean annual temperature is 20 °C. The vegetation is comprised of upper montane rainforest. An approximately 10 m-thick regolith profile (including a zone with corestones at the bottom, a deeply weathered saprolite layer, and 60 cm of soil on top, Fig. 1A) developed over a charnockitic bedrock lithology, with plagioclase (An25), K-feldspar, and quartz as major mineral components and minor amounts of biotite and orthopyroxene. The weathering of the bedrock is initiated along large conjugated fractures dividing the bedrock into blocks. The separated blocks of charnockite weather spheroidally resulting in a transition zone featuring rounded corestones between coherent bedrock and deeply weathered saprolite. Due to the inward advance of weathering, corestones are surrounded by spheroidally spalled “rindlets” (Buss et al., 2008). These spheroidally weathered corestones feature several weathering fronts on a cm- to dm-scale, separating a virtually unweathered core from saprolite-like material in the outer rindlets. Despite humid, tropical climate and high relief the chemical weathering (chemically removed material) and total denudation (sum of chemical weathering and physical