

Climate dependence of feldspar weathering in shale soils along a latitudinal gradient

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Received 16 August 2012; accepted in revised form 2 August 2013; available online 12 August 2013

Abstract

Although regolith, the mantle of physically, chemically, and biologically altered material overlying bedrock, covers much of Earth's continents, the rates and mechanisms of regolith formation are not well quantified. Without this knowledge, predictions of the availability of soil to sustain Earth's growing population are problematic. To quantify the influence of climate on regolith formation, a transect of study sites has been established on the same lithology – Silurian shale – along a climatic gradient in the northern hemisphere as part of the Susquehanna Shale Hills Critical Zone Observatory, Pennsylvania, USA. The climate gradient is bounded by a cold/wet end member in Wales and a warm/wet end member in Puerto Rico; in between, mean annual temperature (MAT) and mean annual precipitation (MAP) increase to the south through New York, Pennsylvania, Virginia, Tennessee and Alabama. The site in Puerto Rico does not lie on the same shale formation as the Appalachian sites but is similar in composition. Soils and rocks were sampled at geomorphologically similar ridgetop sites to compare and model shale weathering along the transect. Focusing on the low-concentration, non-nutrient element Na, we observe that the extent and depth of Na depletion is greater where mean annual temperature (MAT) and precipitation (MAP) are higher. Na depletion, a proxy for feldspar weathering, is the deepest reaction documented in the augerable soil profiles. This may therefore be the reaction that initiates the transformation of high bulk-density bedrock to regolith of low bulk density. Based on the shale chemistry along the transect, the time-integrated Na release rate (Q_{Na}) increases exponentially as a function of MAT and linearly with MAP. NY, the only site with shale-till parent material, is characterized by a Q_{Na} that is 18 times faster than PA, an observation which is attributed to the increased surface area of minerals due to grinding of the glacier and kinetically limited weathering in the north. A calculated apparent Arrhenius-type temperature dependence across the transect (excluding NY) for the dissolution of feldspar (Na depletion) is $99 \pm 15 \text{ kJ mol}^{-1}$, a value similar to field-measured values of the activation energy (14–109 kJ mol^{-1}) or laboratory-measured values of the enthalpy of the albite reaction (79.8 kJ mol^{-1}). Observations from this transect document that weathering losses of Na from Silurian shale can be understood with models of regolith formation based on chemical and physical factors such that weathering progresses from kinetically limited sites (Wales to AL)

Abbreviations: SSHO, Susquehanna Shale Hills Critical Zone Observatory; MAT, mean annual temperature; MAP, mean annual precipitation; LGM, Last Glacial Maximum; SRT, soil residence time; GIS, Geographic Information Systems; NRCS, Natural Resource Conservation Service; ICP-AES, inductively coupled plasma atomic emission spectroscopy; MCL, Materials Characterization Laboratory; LOI, loss on ignition; XRD, X-ray diffraction

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to the transport-limited site in Puerto Rico. Significant advances in our ability to predict regolith formation will be made as we apply more quantitative models to such transect studies on shales and other rocks types.

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1. INTRODUCTION

The Critical Zone, which supports most terrestrial life on Earth, extends from the top of the vegetation canopy to the lower limits of aquifers beneath Earth's continental surfaces (National Research Council, 2001; Brantley et al., 2007). As a central constituent of this zone, soil is the interface between gas and water exchange connecting the atmosphere and aquifers and plays a major role in nutrient cycling that supports ecosystems and humans alike (Amundson et al., 2007). The rate at which soil forms, however, cannot be quantified from soil state factors (Amundson, 2004; Minasny et al., 2008) even though many researchers have sought to quantify physical erosion, pedogenesis and geochemical fluxes as a function of climate and parent material (e.g. Chadwick et al., 1990; Rinaldo et al., 1995; White and Blum, 1995; Steefel and Lichtner, 1998; White et al., 1999; Riebe et al., 2003; Godd ris et al., 2006; Rasmussen et al., 2007; Buss et al., 2008; Dixon et al., 2009a,b; Jin et al., 2010; Maher, 2010; Rasmussen et al., 2011; Moore et al., 2012). Predictions of the long-term availability of soil to sustain agriculture for Earth's growing population are problematic given this lack of knowledge.

Shale is of particular interest because it is a common sedimentary rock at Earth's surface, constituting approximately 25% of mapped bedrock (Amiotte-Suchet et al., 2003) and 51% of the sedimentary lithosphere (Lerman and Wu, 2008). In addition, shale has relatively limited diversity of mineralogy given its origin from previously weathered materials, meaning modeling soil formation on shale may be simpler than such models for crystalline rocks. Previous field studies of shale weathering have largely focused on organic-rich ("black") shale weathering, a small subset of all shales (e.g. Littke et al., 1991; Kolowitz and Berner, 2002; Tuttle and Breit, 2009; Woodruff et al., 2009; Mathur et al., 2012). Laboratory studies of shale dissolution have been performed on both organic-poor and organic-rich shales (Liermann et al., 2011). Likewise, detailed studies of shale weathering are under way at the Susquehanna Shale Hills Critical Zone Observatory (SSHO) in central Pennsylvania, USA (Lin et al., 2006; Jin et al., 2010; Ma et al., 2010; West et al., 2013). The research presented here builds upon the work at SSHO and thus focuses on weathering of organic-poor shale along a climosequence spanning from tropical to high mid-latitude temperate regimes. In this paper we provide the first description of the soils along the climosequence to catalyze efforts to formulate quantitative models of regolith formation.

2. BACKGROUND

The formation of soil has long been considered within the theoretical framework proposed by Jenny (1941), where observed soil properties are a function of climate, landscape

position, organisms, parent material and time. The type of soil and the depth and degree of development are influenced by these five factors. Today, we recognize that the effect of biota must also include anthropogenic influences (Amundson et al., 2007; Wilkinson and McElroy, 2007; Richter, 2007). An environmental gradient approach has often been used by pedologists to isolate the influence of each variable (e.g. Bockheim, 1980; Birkeland, 1999; Williams et al., 2010). For the investigation reported here, study sites were carefully selected along a 34° latitudinal climate gradient (18°N–52°N) primarily on Rose Hill shale within the Appalachian Mountains, holding state factors other than climate as constant as possible (discussed further below). Additional sites beyond the Appalachian Mountains include a similar shale in the tropics (Puerto Rico) and a shale in Wales, United Kingdom that is stratigraphically and geochemically equivalent to the Rose Hill Formation in the United States. Sites were chosen only in regimes where precipitation is greater than evapotranspiration and strictly from ridge top positions. All sites therefore experience net water flow into the subsurface that approaches vertical without the complexity of downslope transport of water or sediments (Jin et al., 2010). In summary, the variables of lithology, vegetation, erosion rate and human disturbance are held relatively constant while temperature, precipitation, relief and bedrock orientation vary along the transect.

One method for determining the extent of weathering is to measure the mass loss from an observed soil profile relative to the original concentrations in unweathered rock (April et al., 1986; Brimhall and Dietrich, 1987; Brimhall et al., 1992; White et al., 1998; Brantley et al., 2008; Brantley and White, 2009; Brantley and Lebedeva, 2011). Using this approach, relative changes in the concentration of the mobile elements which are removed from the soil profile during weathering are compared to the concentration of an immobile element not involved in weathering. This approach has been used to estimate field weathering extents of the dominant crystalline rocks (granite and basalt) as well as shale (e.g. April et al., 1986; White et al., 2001; Chadwick et al., 2003; Jin et al., 2010). The method is contingent upon the proper identification of the parent material. Once the mass loss of an element has been calculated, an estimation of the duration of weathering allows determination of a weathering rate (White, 2002; Brantley and White, 2009; Brantley and Lebedeva, 2011). Depletion of an individual element can be used as a proxy for the extent of mineral reactions provided the element is found in one mineral. For example, Na loss is often attributed to feldspar weathering (White and Brantley, 2003) and Mg loss can indicate biotite, hornblende, or chlorite weathering (Luce et al., 1972; Buss et al., 2008; Jin et al., 2010). Such methods have also been used across large latitudinal gradients. For example, elemental release rates have been used to

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