

**Data Repository Item:****Mineral persistence:**

Figure 1 includes soils classified as “residual soils” (little loss by erosion), “steady state soils” (equal rates of loss and formation of regolith), or “degrading soils” (regolith loss rate > formation rate). A mineral life-time or persistence time is therefore defined as i) the duration of time a mineral was exposed to cosmogenic rays in the upper meter of regolith (where a cosmogenic age is available), or ii) the maximum estimated time of exposure to weathering based on landform arguments (where cosmogenic ages are unavailable). Landform ages are estimated based upon emplacement of a lava flow, time of last glaciation, or time of marine planation. Where cosmogenic dates are available and the soil is not a steady state soil, persistence ages are  $\leq$  landform ages and represent the residence time for the mineral within the soil depth interval affected by cosmogenic rays. All phase compositions and climates available are included in this figure. All laboratory-based rates in Figure 1 are from Brantley (2003) except for basalt glass, which is calculated from the basalt glass rate equation in Bandstra and Brantley (in press), and rhyolitic glass, which is estimated to be 9x slower than basalt based on experiments by Wolff-Boenisch et al. (2004) at pH = 4. Field data are compiled from the literature (Dickson and Crocker, 1954; Hay, 1960; Hendricks and Whittig, 1968; Locke, 1979; Ruxton, 1988; Veldkamp et al., 1990; Bain et al., 1993; Wasklewicz, 1994; Bockheim et al., 1996; White, 1996; Darmody and Thorn, 1997; Vitousek et al., 1997; Dubroeuq et al., 1998; Karrat et al., 1998; White et al., 1998; Burkins et al., 1999; Nieuwenhuysen et al., 2000; Oguchi, 2001; Pinney et al., 2002; Yokoyama and Banfield,

2002; Egli et al., 2003; Sedov et al., 2003; Chesworth et al., 2004; Mikesell et al., 2004; Stephens and Hering, 2004; Buss, 2006).

### **Mineral dissolution rates as a function of pH:**

The fayalite rate in Figure 2 is estimated assuming that fayalite dissolves 10x faster than forsterite (Bandstra and Brantley, in press) as documented in the literature at acidic to neutral pH (Wogelius and Walther, 1992). The bytownite dissolution rate is derived from published parameters (Blum and Stillings, 1995; Brantley, 2003; Bandstra and Brantley, in press).

### **Insights from terrestrial basalt weathering rinds:**

Weathering rinds are chemically altered surfaces on parent rock (Figure 3). We consider here rinds on Svalbard basalt that show partial loss of Na-containing glass with little secondary mineral precipitation or loss of other phases (Figure 3b) and rinds on Costa Rica basaltic material that show complete loss of Mg-pyroxene and Na-plagioclase and replacement by secondary minerals (Sak et al., 2004). To account for non-isovolumetric weathering or mobility of multiple elements, concentrations must be normalized to an immobile element (Anderson et al., 2002) (Figure 4),

$$\tau_{i,j} = \frac{C_{j,w}}{C_{j,p}} \frac{C_{i,p}}{C_{i,w}} - 1, \quad (1)$$

where  $\tau_{i,j}$  is the fraction of mobile element or mineral  $j$  lost or gained assuming that element or mineral  $i$  is immobile ( $w$  and  $p$  refer to weathered and parent material respectively) and  $C$  is the concentration of the immobile and mobile elements in the parent and weathered materials. Titanium, a relatively abundant element, is often

considered immobile because it is generally mobile only in concentrated acids or when found in highly soluble Ti minerals. Depletions in  $\tau_{i,j}$  can document reaction fronts where weathering fluids, undersaturated with respect to primary minerals at the outer surface, have dissolved minerals throughout the rind until chemical equilibrium is approached at depth (Lichtner, 1988). Extensive weathering (Figure 4a) reduces  $\tau_{i,j}$  to -1 (complete depletion), whereas less intensive weathering or high erosion of surfaces show  $-1 < \tau_{i,j} < 0$ .

From models in the literature, the thickness of the reaction front for a diffusion-dominated weathering rind (defined as the portion of the  $\tau$ -depth curve where  $\tau_{i,j}$  changes from 0 (parent) to the lowest value in the profile) represents the square root of the product of porosity ( $\phi$ ) and diffusivity ( $D$ ) divided by the product of reaction rate constant  $k$  and mineral-water interfacial area ( $A$ ):

$$l = \left( \frac{\phi D}{kA} \right)^{1/2} \quad (2)$$

Strictly, this equation only holds for weathering of a single-component, single-phase system characterized by linear kinetics (Lichtner, 1988). For multi-component, multi-phase systems, this equation cannot strictly be applied; nonetheless, steeper fronts represent slower weathering rates relative to solute transport as compared to shallow fronts.

The Costa Rican samples are clasts weathered at MAT 27.3°C and MAP 3085 mm in an alluvial terrace deposited 35ka ago (Sak et al., 2004), while the Svalbard samples are Quaternary basalt lavas weathered *in situ* since glaciation last stripped the

rock surfaces ~10ka ago. Weathering is conceptualized here as mediated by diffusive transport through fully wetting pore fluids (Sak et al., 2004).

#### **Weathering rind analysis for Humphrey:**

The elemental profile through Humphrey is normalized to Ti as an assumed immobile element: the Ti concentrations in Humphrey are the same within uncertainty throughout the profile. The total depth of the RAT grind is from Arvidson (2006), and the first RAT grind is assumed to be 50% of the depth of the total RAT grind. The elemental compositions are from Gellert et al. (2006). The Mars profile was first corrected for dust by assuming that the brushed surface of the rock (Gellert et al., 2006) contained the same percentage of sulfur as the deepest parent material, and assuming that all sulfur present in greater amounts is due to dust. Dust was assumed to be compositionally identical to the average of the two unbrushed surface measurements on Humphrey.

#### **Reactive transport modeling:**

pH and aqueous concentrations for Costa Rica and Svalbard are based on measured pore water concentrations from the sample sites. pH was measured upon addition of 2.5 ml of deionized water to 1 g of soil for both Costa Rica and Svalbard (Brady and Weil, 2002). Pore water concentrations were measured on porefluids collected after equilibrating (6 hrs at ~4°C) 5 mL of deionized water with 15 g of field-wet samples for both Costa Rica and Svalbard. The slurry was centrifuged at 4500 rpm for 35 minutes at 5°C, filtered through a 0.2 µm filter, acidified, and measured on a Finnigan Element I Inductively Coupled Plasma Mass Spectrometer. In CrunchFlow, the

outer surface of each rock was assumed to be bathed in a constant concentration pore fluid of appropriate pH.

We have used surface area as a fitting term in Costa Rica, Svalbard and Mars; however, the fitted values for surface area were all within a factor of ~100 of geometric, suggesting our assumed values are defensible. The surface area of the glass grains in Svalbard can be estimated by examining the reaction front in the basalt. Glass occurs interstitially in the rock; approximate dimensions of interstices are on the order of 10  $\mu\text{m}$ . However, little of this glass has interacted with porefluid because little porosity exists. In contrast, pores delineate aggregates of glass + mineral matrix that we define as “apparent grains”. We measured the approximate diameter (~40 $\mu\text{m}$ ) of these apparent grains and then calculated a geometric surface area.

Laboratory dissolution rate constants are generally normalized by BET surface areas, and therefore surface area for natural samples is typically estimated from the geometric surface area multiplied by a roughness value (= ratio of BET surface area to geometric surface area). However, reactive surface area may only equal 0.1 to 10% of the BET surface area (Velbel, 1993).

*Appendix Table DR 1: Modeling parameters*

	Svalbard	Mars	Costa Rica
Time: (years)	2500 (observed <sup>1</sup> )	22000 (model inference)	35000 (observed)
Temperature: (°C)	2 <sup>2</sup>	1 <sup>3</sup>	25 <sup>4</sup>
pH:	8.65	4	5
Reacting fluid concentrations	measured	(Hedin et al., 2003)	measured
Hydrologic saturation	100%	100%	100%
Primary silicates	Glass	olivine	Plagioclase pyroxene
Secondary minerals	none	none	Kaolinite, Fe(OH) <sub>3</sub> , gibbsite, siderite

Glass surface area (m <sup>2</sup> /g)	0.05	NA	NA
Olivine surface area (m <sup>2</sup> /g)	NA	0.00001	NA
Plagioclase surface area(m <sup>2</sup> /g)	NA	NA	0.0005
Pyroxene surface area (m <sup>2</sup> /g)	NA	NA	0.00039
Tortuosity:	0.000001 <sup>5</sup>	0.000001 <sup>5</sup>	Archie's law (cementation exponent of 2.5)
Dissolution constants:	(Bandstra and Brantley, in press) <sup>6</sup>	(Bandstra and Brantley, in press)	(White and Brantley, 1995)

<sup>1</sup> The age of the landform (10ka since deglaciation) multiplied by the approximately one quarter of the year that the landform is not frozen.

<sup>2</sup> Assumed average temperature above freezing

<sup>3</sup> Low model temperature above zero

<sup>4</sup> Observed temperature =27.3

<sup>5</sup> Tortuosity is based on laboratory-measured values for basalt (Sato et al., 1997).

<sup>6</sup> Natural glasses vary in composition, including the glass compositions we measured in Svalbard, which vary from approximately basaltic to approximately andesitic. Glass dissolution rates vary as a function of composition (Hamilton, 1999; Wolff-Boenisch et al., 2004). In this work we use the dissolution rates of basalt glass as fit by Bandstra and Brantley (2007), recognizing that these rates may vary as a function of composition and that this therefore represents a simplification.

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