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Climatically controlled delivery and retention of meteoric ^{10}Be in soils

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ABSTRACT

Meteoric ^{10}Be (^{10}Be$_m$) is a widely used tracer of soil processes in terrestrial ecosystems, but complexity surrounding its delivery and retention in soils is frequently oversimplified. We measured ^{10}Be$_m$ in ten soil profiles sampled on 150 ka lava flows of Kohala, Hawaii. The sampled soils receive annual rainfall of 160–3000 mm, and exhibit strong gradients in chemical properties. Below ~1400 mm rainfall, ^{10}Be$_m$ inventories in the upper 1–2 m of soils span ~37–270 × 10^9 atom cm^{-2} and increase linearly with rainfall, consistent with precipitation-driven depositional fluxes and the retention of ^{10}Be$_m$ in clay-rich soil horizons. ^{10}Be$_m$-derived ages, based on flux estimates and measured inventories, are uniform and consistent with the known substrate age (150 ka) at these sites. However, nuclide inventories dramatically decrease for soils at >1400 mm rainfall, where water balance changes from negative to positive. This climate-driven threshold is associated with ^{10}Be$_m$ mobility, high pore-water volumes, low base cation saturation, and high exchangeable Al. Though rainfall-mediated delivery of ^{10}Be$_m$ to soil is relatively predictable, its mobility and retention are strongly constrained by water-mediated ion exchange properties. Our results indicate meteoric ^{10}Be may not be a robust tracer of soil age in systems with high rainfall and weathering intensity.

INTRODUCTION

Meteoric ^{10}Be (^{10}Be$_m$) is a promising tracer of soil residence times and movement (Monaghan et al., 1983), though uncertainties in its delivery to and retention in soil have limited its widespread use. Its widely used twin, in situ ^{10}Be, is produced by cosmic rays in the lattices of mineral grains, while the meteoric variety is produced in the upper atmosphere and deposited via both dry and wet processes (Field et al., 2006). Beryllium has a high partition coefficient in most environmental settings, binding strongly to soils and sediments at Earth's surface, and has a long residence in natural materials (Pavich et al., 1986). Given assumptions about its production and delivery, ^{10}Be$_m$ is used to calculate soil and surface ages, rates of erosion and weathering, and soil movement across a landscape (Graly et al., 2010; West et al., 2013; Willenbring and von Blanckenburg, 2010).

One limitation to the use of ^{10}Be$_m$ is uncertainty about its delivery from the atmosphere to land surface (Reusser et al., 2010; Ouimet et al., 2015). The majority of ^{10}Be$_m$ is delivered as wet deposition, though dust contributions can be non-trivial (e.g., Brown et al., 1989; Ouimet et al., 2015). Atmospheric ^{10}Be$_m$ scavenging is complex (Field et al., 2006; Monaghan et al., 1986; Willenbring and von Blanckenburg, 2010). Regardless, models of global ^{10}Be$_m$ delivery generally show annual deposition rates are high in regions with high precipitation, and limited measurements of atmospheric ^{10}Be$_m$ fluxes suggest strong variation with precipitation (Graly et al., 2011, Graham et al., 2003).

A second limitation in the use of ^{10}Be$_m$ concerns its behavior in terrestrial systems (Bacon et al., 2012). The assumption of ^{10}Be$_m$ retentivity in soils is required to derive soil ages and erosion rates (Bacon et al., 2012; Maher and von Blanckenburg, 2016; Pavich et al., 1986). Mobility of ^{10}Be$_m$ within soil profiles has been previously suggested in soil chronosequence studies (e.g., Monaghan et al., 1983; Pavich et al., 1984; Barg et al., 1997), and by comparison with the behavior of rock derived ^{9}Be (Bacon et al., 2012). Laboratory experiments indicate organic C speciation and pH influence Be binding and mobility in soils and sediments (You et al., 1989; Boschi and Willenbring, 2016). However, there are few observations of the conditions governing ^{10}Be mobility directly within soils.

Here, we evaluate ^{10}Be$_m$ delivery and retention along a well characterized soil climosequence on the leeward slope of Kohala, Hawaii. We quantify ^{10}Be$_m$ in soils formed on constructional lava flows of the same age but under strongly differing annual rainfall. These data improve understanding of ^{10}Be$_m$ systematics and retentivity in soil, and provide new insight into the limitations of this isotope system.

APPROACH

The island of Hawaii is an ideal system to quantify the effect of rainfall on soil processes; the uniformity of parent material composition and age make rainfall the dominant variable affecting soil properties on the south slope of the Kohala Peninsula (Chadwick et al., 2003). Northeast trade winds bring moist air to windward slopes, creating a leeward rain shadow with a strong and stable climate gradient. Abundant prior work across this system has characterized soil and elemental geochemistry (Chadwick et al., 2003), weathering intensity and nutrient biocycling (Bullen and Chadwick, 2016), and dust fluxes (Porder et al., 2007).

Ten sites were sampled along a 12 km transect on leeward Kohala (Fig. 1). Annual precipitation at the sites varies from 160 to 3000 mm (Chadwick et al., 2003). Sampling sites were selected to minimize local erosion/deposition effects, and soil geochemical properties for the samples are described in Chadwick et al. (2003). Soil profiles were sampled by horizon to saprolite or rock, and dried and sieved. Total rock fragments (>2 mm) were used for volume/mass corrections assuming they contained negligible ^{10}Be$_m$. The <2 mm fraction was ground with a mortar and pestle. A 1 g aliquot was subsampled from the representative soil powder, spiked with 1mg of ^{9}Be, and processed through sequential leaches, ion-exchange columns, and precipitation steps to extract Be following the methods

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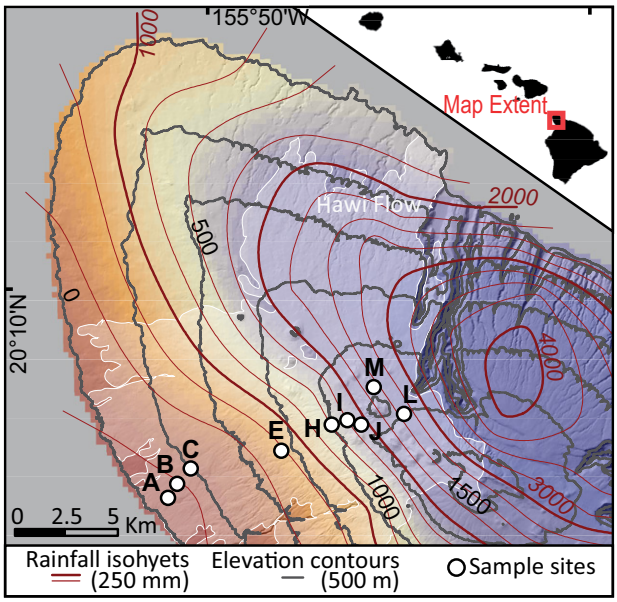

Figure 1. Sample sites on the Kohala climate gradient of Hawaii are located within the Hawi basalt flow (150 ka; transparency) and span 160–3000 mm mean annual precipitation (Giambelluca et al., 2013). Site B' is ~20 m from Site B.

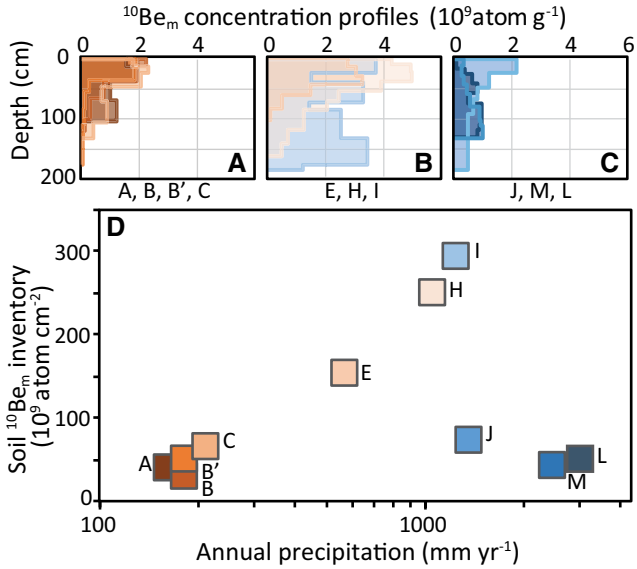

Figure 2. Meteoric ^{10}Be ($^{10}Be_m$) is concentrated at the surface of driest profiles (a), and increasingly distributed to depth (b) with increasing precipitation. Wettest profiles (c) show low $^{10}Be_m$ throughout sample depths. Soil $^{10}Be_m$ inventories of soils (upper 1–2 m) (d) increase linearly with annual precipitation and then decrease precipitously above a rainfall threshold of ~1400 mm. Color shading is consistent throughout figures.

outlined by Brown et al. (1981). Be was combusted at 550 °C to form BeO, mixed with Nb, and packed into targets for accelerator mass spectrometry at the Lawrence Livermore National Lab (LLNL; California, USA) facility. Reported ^{10}Be /^{9}Be ratios were originally normalized to standard LLNL 10000 ($^{10}Be/^{9}Be = 1.00 \times 10^{-11}$), and data were recalibrated to the revised ICN standard following Nishiizumi et al. (2007). $^{10}Be_m$ inventories are calculated as the summed product of horizon thickness, soil density, and ^{10}Be concentration, and are decay corrected using a half-life of 1.36 ma and parent material age of 150 ka.

RESULTS AND DISCUSSION

Meteoric ^{10}Be in Kohala Soils

Meteoric ^{10}Be concentrations in the Kohala soils vary strongly with rainfall and depth (Fig. 2). In sites receiving <500 mm rainfall, $^{10}Be_m$ declines from near-surface maxima of ~2×10^9 atom g^{-1} to negligible values at ~100 cm depth (Fig. 2a), whereas intermediate rainfall sites (500–1300 mm) contain 1.5–2 times greater $^{10}Be_m$ concentrations near the surface and ^{10}Be is distributed to nearly 200 cm depth (Fig. 2b). At high rainfall (>1400 mm), sampled soils contain considerably less $^{10}Be_m$ (calculated as inventories) and the two wettest sites show greater concentrations deep in the profiles compared with the surface (Fig. 2c). Down-profile redistribution of $^{10}Be_m$ has been noted in other soils, particularly those with subsurface accumulations of clay minerals (Pavich et al., 1986; Bacon et al., 2012). Observed changes in $^{10}Be_m$ profiles of upper soil horizons are accompanied by inventories (Fig. 2d) that increase linearly with annual rainfall up to 1400 mm ($r^2 = 0.99$). For sites receiving >1400 mm, inventories drop nearly an order of magnitude.

We derive soil 'meteoric ^{10}Be ages' from decay-corrected $^{10}Be_m$ inventories and estimates of atmospheric flux at each sample location. Direct measurements of $^{10}Be_m$ in rainfall at Mauna Loa (Brown et al., 1989) give modern rainfall concentrations of $16.9 \pm 3.5 \times 10^3$ atom mL^{-1}. Applying this concentration across the rainfall gradient yields ages that fall into two clear groups (Fig. 3). Soils with annual rainfall <1400 mm show calculated ages of 145 ± 30 ka, similar to the 150 ka age of the parent Hawi basalt a'a flows. Inferred ages drop roughly an order of magnitude to 17 ± 13 ka across the threshold at >1400 mm rainfall.

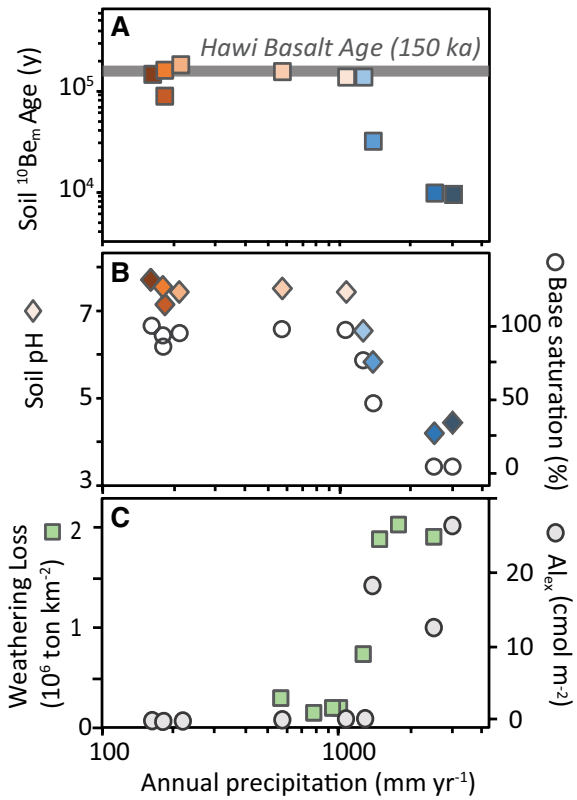

Figure 3. (a) Soil meteoric ^{10}Be ($^{10}Be_m$) ages are consistent with the age of parent material at low annual rainfall (<1400 mm). Lower inferred ages at high rainfall are due to $^{10}Be_m$ loss from soils and coincide with pedogenic thresholds where (b) base saturation and soil pH decrease markedly (Chadwick et al., 2003) as total weathering increases (Porder et al., 2007). (c) Extractable Al likely outcompetes Be for exchange sites, increasing ^{10}Be mobility.

Geochemistry and Retention of ^{10}Be

The sudden decrease in $^{10}Be_m$ inventories of sampled soils above 1400 mm rainfall is notable considering the uniform age and composition of the parent material. This transition reflects one from Be retention to Be mobility across a previously identified threshold in soil chemical properties (Chadwick and Chorover, 2001).The threshold at 1400 mm is coincident with the change in annual water availability from deficit to surplus (in excess of potential evapotranspiration), and is associated with a strong decline in base saturation (Chadwick et al., 2003), the percent of available exchange sites occupied by major non-hydrolyzing cations compared with Al(OH)x. Depth-averaged base saturation declines from ~80%–5% across this threshold, and soil pH decreases from 7.5 to less than 5.0 (Chadwick et al., 2003; Fig. 3b). Total weathering losses increase across this threshold by nearly an order of magnitude (Porder et al., 2007; Fig. 3c). Therefore, the observed decrease in $^{10}Be_m$ inventories at rainfall >1400 mm is likely explained by the strong weathering threshold and the physico-chemical processes that alter clays and mediate sorption of cations to surfaces of soil colloids.

That $^{10}Be_m$ is lost violates commonly held assumptions regarding retention in soils (e.g., Pavich et al., 1984; West et al., 2013). At near neutral pH, Be^{2+} has high partition coefficients, indicating strong attachment onto soil particles (Kd >10^5 in clays, oxyhydroxides, and clastic sediments; You et al., 1989). Therefore, $^{10}Be_m$ delivered in rainfall is likely retained in soil horizons with sufficient cation exchange capacity (Pavich et al., 1986). However, sorption and desorption experiments (Aldahan et al., 1999; You et al., 1989) indicate that Be retention on mineral surfaces changes markedly below solution pH of 7, such that partition coefficients decrease by several orders of magnitude (see the GSA Data Repository[1]). Furthermore, in lab experiments from Boschi and Willenbring (2016) reduction of solution pH from 6 to 4 resulted in up to 97% Be desorption from some clays.

Laboratory findings are consistent with the observed decrease in $^{10}Be_m$ soil inventories (and inferred losses) as soil pH and base saturation decrease (Figs. 2 and 3b). But, even at reduced pH in surface soils, we expect retention of Be lower in the profile, or to be tightly held within authigenic clays and Al- and Fe-hydroxides (Barg et al., 1997). While cation exchange capacity varies with rainfall (Chadwick et al. 2003), it is not so low to directly limit ion exchange and potential ^{10}Be sorption, and all soils are dominated by clay minerals with high concentrations of short-range order (SRO) clay minerals (e.g., allophane and ferrihydrite). Thus, leached species are lost despite abundant exchange sites. Loss of Be from the upper soils therefore likely reflects multiple processes beyond simple desorption (Graly et al., 2010), including leaching of SRO minerals and/or translocation on soil colloids (Bacon et al., 2012). The decrease in ^{10}Be inventories occurs at the transition to positive water balance and is associated with increasing water volume passage through the soil column. Potential throughflow volumes and surplus water available for weathering reactions increase rapidly with rainfall beyond the 1400 mm threshold (Chadwick et al., 2003). Thus, leached species are lost quickly from upper soil horizons with pore water flow, such that exchange sites cannot re-adsorb ions.

Cation exchange also plays a role in observed Be losses. Extractable Al and Be are highly correlated in several studies (Graly et al., 2010; Jungers et al., 2009; Monaghan et al., 1983) indicating similar behavior of these cations. Thus, $^{10}Be_m$ could be mobilized similar to Fe and Al in soil profiles, especially in association with certain organic ligands (Monaghan et al., 1983). In the soils observed here, increased organic matter content and organic complexation of these metals may facilitate removal from the mineral ion-exchange (Kurtz et al., 2000) and prior work noted higher concentrations of crystalline aluminosilicates (halloysite) and metal oxides (hematite and goethite) in drier soils compared to sites above the rainfall threshold (Chadwick et al. 2003). The decline of $^{10}Be_m$ concentrations below those predicted from meteoric fluxes suggests that trivalent Al released by weathering (Fig. 3c) outcompetes divalent Be and its isotopes for exchange sites under decreasing pH values and increasing pore water passage.

Meteoric ^{10}Be Delivery in Rainfall and Dust

Uncertainties in $^{10}Be_m$ flux and its controls have been a major limitation in applications to soil chronology studies. Direct measurements of $^{10}Be_m$ in rainfall at Mauna Loa (Brown et al., 1989; 16.9 ± 3.5 × 10^3 atom mL^{-1}), applied to the range of annual rainfall across sites A through I, yield estimated short-term $^{10}Be_m$ wet-deposition fluxes between 270 and 2130 × 10^3 atom cm^{-2} y^{-1}. Long-term fluxes based on $^{10}Be_m$ inventories and the parent material age are similar (270–1940 × 10^3 atom cm^{-2} y^{-1}), and consistent with $^{10}Be_m$ concentrations of rainfall (averaging 15.2 × 10^3 atom mL^{-1} over soil residence). The similarity in patterns of measured short-term (Brown et al., 1989) and modeled long-term $^{10}Be_m$ fluxes (Fig. 3a) suggests that the structure of the climate gradient has largely persisted over 150 ka of soil development.

The ability to compare both long-term and short-term flux estimates is rare (e.g., Ouimet et al., 2015). While models of $^{10}Be_m$ flux provide useful estimates at a global scale (Heikkilä and von Blanckenburg, 2015; Graly et al., 2011), they differ from measured values in Hawaii by up to an order of magnitude (Fig. 4), resulting in equally variable age estimates. $^{10}Be_m$ delivery is highly correlated with rainfall (Brown et al. 1989) and orography, resulting $^{10}Be_m$ flux being strongly site specific.

Dust also provides a source of $^{10}Be_m$ to soils (Ouimet et al., 2015), and has been noted as a significant source of Sr and other elements to parts of Hawaii. Across the climate gradient, dust deposition ranges from 20 to 130 g cm^{-2} ky^{-1} based on quartz and Nd-isotope tracers (Kurtz et al., 2001; Porder et al., 2007). This dust is derived from loess terrain of eastern Asia where the background $^{10}Be_m$ is ~2.8 × 10^8 atom g^{-1} (Shen et al., 2010). Applying this estimate to measured $^{10}Be_m$ concentrations in soils, the $^{10}Be_m$ flux with dust ranges between 4.9 and 30.4 × 10^3 atom cm^{-2} y^{-1},

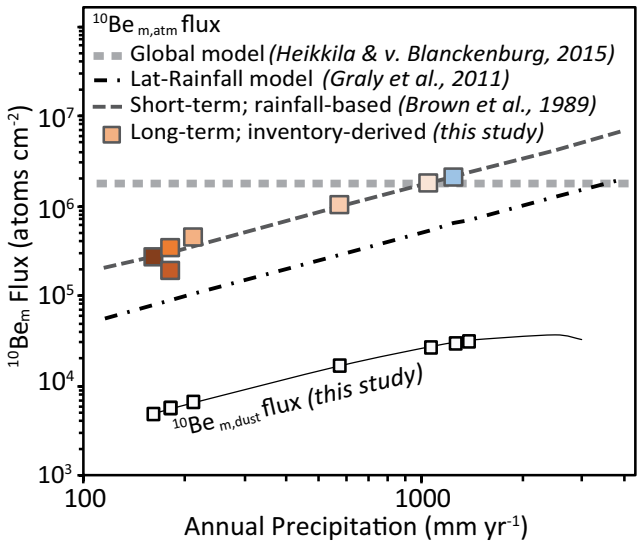

Figure 4. Long-term atmospheric meteoric ^{10}Be ($^{10}Be_m$) fluxes, based on parent material age and soil inventories of sites <1400 mm rainfall, match short-term measurements in modern rainfall (Brown et al., 1989). These fluxes differ from those derived via empirical models (Graly et al., 2011) and show local variation not representable in coarse global models of production and delivery (Heikkilä and von Blanckenburg, 2015). $^{10}Be_m$ fluxes in dust, based on dust deposition rates (Porder et al., 2007) and dust $^{10}Be_m$ (Shen et al., 2010), constitute a small fraction of total $^{10}Be_m$ flux (< 1.5%).

[1]GSA Data Repository item 2018327, supplementary data, is available online at http://www.geosociety.org/datarepository/2018/ or on request from editing@geosociety.org.

and total $^{10}Be_m$ delivered over the 150 ka of soil residence spans 0.7–4.6 × 10^9 atom cm^{-2}. These fluxes represent 1.6%–2.1% of estimated inventories (excluding sites at high rainfall). Though dust may provide important nutrient fluxes to old soils in Hawaii, it is a trivial mode of $^{10}Be_m$ delivery to soils in this system, even over long timescales.

CONCLUSIONS

Meteoric ^{10}Be data from soils across Kohala, Hawaii provide several new insights into the delivery and retention of $^{10}Be_m$ and similar tracers in terrestrial environments. Firstly, our results show that a transition from negative to positive water balance across the rainfall gradient marks a threshold for Be desorption and mobility in upper soil horizons where $^{10}Be_m$ is delivered. Soils dominated by low leaching intensities can retain $^{10}Be_m$ and its isotopic record of soil residence times, erosion rates, and paleo-environmental conditions. We find $^{10}Be_m$ loss is associated with a combination of soil physico-chemical conditions, including high throughflow, low pH and base saturation, and mobilization of other cations and metals. Considering that moisture-driven weathering thresholds can establish rapidly (Vitousek et al., 2016; Dixon et al., 2016), transitions to wetter climates for short timescales relative to that of soil formation may result in losses of these meteoric tracers from soils.

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