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2 **Surface ages and weathering rates from ^{10}Be (meteoric) and $^{10}\text{Be}/^9\text{Be}$: insights**
3 **from differential mass balance and reactive transport modeling**

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Abstract

To reconstruct the dynamic evolution of weathered material at the Earth's surface requires techniques that record the fluxes of material into and out of the weathering zone. Cosmogenic isotopes, formed *in situ* within the Earth's surface or in the atmosphere ("meteoric") are important chronometers for both time and fluxes. Meteoric ^{10}Be has been used to evaluate surface ages, soil residence times and rates of sediment transport; however, the meteoric ^{10}Be dating tool relies on complete retention of ^{10}Be on mineral surfaces within the weathered regolith, an assumption that will not be valid where soil pH is low or mineral surface area available for sorption is lacking. Here we evaluate whether the stable isotope of ^9Be that is released upon weathering ("reactive" Be) can be used to correct for ^{10}Be mobility. This approach relies on the inventory of reactive ^9Be in the weathering zone as a measure of the mass depletion, whereas the ^{10}Be inventory is a function of age. To explore the spatial and temporal relationship between the Be isotopes, we use a multi-component reactive transport model framework to evaluate a set of differential mass balance equations for the $^{10}\text{Be}/^9\text{Be}$ composition of the bulk solid and sorbed reservoirs on non-eroding surfaces. We first find that the $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ is inversely proportional to the long-term chemical weathering rate, even in cases of incomplete Be retention. Second, we find that coupling ^{10}Be and ^9Be to provide a tool for surface age dating (^{10}Be) is complex because incomplete retention affects each isotope to a different extent. Beryllium-10 is supplied at the surface of soil, where sorption onto secondary minerals is more extensive, while ^9Be is released from primary minerals at the weathering front where secondary minerals are less abundant. If ^9Be loss is less than about 15%, we find ^9Be inventories can be used to reliably correct ^{10}Be inventories to within 10%. The error associated with the correction also increases with increasing age, and thus the correction is more favorable for younger surfaces. Over longer timescales, the error on the age introduced by the loss correction using ^9Be is magnified, whereas the error associated with uncorrected ages decreases because the ^{10}Be is more strongly retained as chemical weathering progresses. We also find that the reactive transport model confirms a set of simple mass balance descriptions, providing guidance for future data collection. Collectively, the modeling results suggest that measurement of Be isotopes in weathered regolith provides a unique approach to determine both surface ages and chemical weathering rates.

Introduction

Cosmogenic isotopes, produced either in the atmosphere (*meteoric*) or within earth materials at or beneath the surface (*in situ*) have long served as chronometers for both time and fluxes (von Blanckenburg and Willenbring, 2014), allowing the development of conceptual models that allow us to quantify the mechanics of landscapes (Gosse and Phillips, 2001; Granger et al., 2013; Lal, 1988; McHargue and Damon, 1991; Nishizumi et al., 1993). *In situ* cosmogenic ^{10}Be (half-life of 1.39 Myr) is a well-established approach for dating surficial deposits (Nishiizumi et al., 1991; Repka et al., 1997) and determining local and catchment-scale denudation rates (Granger et al., 1996; Kirchner et al., 2001; Nishizumi et al., 1993; Portenga and Bierman, 2011; Schaller et al., 2001). The more abundant variety of Be, meteoric ^{10}Be , has received less attention, in part because of concerns about the mobility of ^{10}Be in the weathering zone (Barg et al., 1997; Monaghan et al., 1983) and uncertainties in the flux of ^{10}Be to the Earth's surface (McHargue and Damon, 1991; Reusser et al., 2010). Nevertheless, the meteoric variety of ^{10}Be (discussed exclusively hereafter) has been used to evaluate surface ages (Duhnforth et al., 2012; Egli et al., 2010), soil residence times (Bacon et al., 2012; Barg et al., 1997; Brown et al., 1992; Pavich et al., 1986) and rates of sediment transport (Brown, 1987; Campforts et al., 2016; Mackey et al., 2009; McKean et al., 1993; West et al., 2014; West et al., 2013). The basis for these approaches is that ^{10}Be formed in the atmosphere is strongly particle reactive, adhering to aerosols that are subsequently deposited to the surface of the weathering zone (McHargue and Damon, 1991; Willenbring and von Blanckenburg, 2010b). If the deposition rate to the land surface ($F^{10\text{Be}}$, $[\text{mol}/\text{m}^2/\text{yr}]$) is known, the inventory of ^{10}Be ($I_{10\text{Be},\text{reac}}$, $[\text{mol}/\text{m}^2]$) is then a function of time (t), radioactive decay (λ , $[\text{yr}^{-1}]$) and loss due to chemical and/or physical processes characterized by a rate constant (L_{10} , $[\text{yr}^{-1}]$):

$$I_{10\text{Be},\text{reac}} = \frac{F^{10\text{Be}}}{\lambda + L_{10}} \left[1 - e^{-(\lambda + L_{10})t} \right]. \quad (1)$$

If there is no loss of ^{10}Be , then the inventory can be used to evaluate surface ages (*e.g.*, Duhnforth et al., 2012; Egli et al., 2010):

$$t = -\frac{1}{\lambda} \ln \left[1 - \frac{I_{10\text{Be},\text{reac}}}{F^{10\text{Be}}} \right]. \quad (2)$$

A key consideration is thus the mobility of ^{10}Be in the weathering zone under a range of conditions.

In order to evaluate and potentially correct for ^{10}Be mobility, one approach is to use the stable isotope, ^9Be . Beryllium-9 is present in ppm concentrations in silicate minerals (von Blanckenburg et al., 2012). The portion of ^9Be that is released upon weathering may mix with meteoric ^{10}Be and sorb to mineral surfaces or be incorporated into secondary phases, resulting in a “reactive” Be pool. The isotopic composition of this reactive Be pool, $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$, reflects the fluxes of both isotopes (von Blanckenburg et al., 2012). It has been shown that in river sediment, unlike ^{10}Be concentrations, the $^{10}\text{Be}/^9\text{Be}$ ratio does not depend on retention or particle size of sediment (von Blanckenburg et al., 2012; Wittmann et al., 2012). Whereas the reactive ^{10}Be concentration can be used to determine erosion rates, the $^{10}\text{Be}/^9\text{Be}$ ratio can be used to infer denudation rates (the sum of erosion and weathering) from soil profiles (Barg et al., 1997), river sediments (von Blanckenburg et al., 2012; Wittmann et al., 2015) and even seawater or seafloor sediment (von Blanckenburg and Bouchez, 2014; von Blanckenburg et al., 2015; Willenbring and von Blanckenburg, 2010a). In soil and weathering profiles, one possibility is to calculate the loss of ^{10}Be using ^9Be mobilized from weathering (Bacon et al., 2012). However, ^{10}Be is supplied at the top of the weathering zone where secondary minerals with high surface areas that act as sorbents are generally more available, whereas ^9Be is released from primary minerals at the weathering front where secondary minerals are less abundant. The spatial separation may result in different retention of the two isotope species (Barg et al., 1997). Such spatial separation may be further exacerbated by biogeochemical gradients within soils. For example, Be tends to sorb strongly to mineral surfaces at $\text{pH} > 5$, and is thus concentrated in clay horizons where the amount of available surface area is high (Jagercikova et al., 2015; Wyshnytzky et al., 2015). At the same time, the formation of organo-beryllium complexes and/or low soil pHs can result in incomplete retention or differential mobility as a function of depth (Barg et al., 1997; Kaste and Baskaran, 2011).

In addition to evaluating ^{10}Be loss for age dating applications, another potential application of the $^{10}\text{Be}/^9\text{Be}$ approach is to determine long-term chemical weathering rates. This approach assumes that the inventory of ^9Be in the weathering zone relative to the parent material ^9Be reflects the mass depletion (ΔM). In contrast, the ^{10}Be inventory is a function of age, or for eroding surfaces, the erosion rate, and as such constrains the soil residence time ($\Delta \tau$). Under the assumption of steady-state regolith thickness, von Blanckenburg et al. (2012) have used the contrasting behavior of the two isotopes to infer denudation rates from the $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$. In the

104 absence of appreciable erosion, the $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ should be inversely proportional to the long-
105 term chemical weathering rate $(\Delta M/\Delta \tau)$. Given the difficulty associated with quantifying long-
106 term weathering rates, particularly in erosional environments, the $^{10}\text{Be}/^9\text{Be}$ approach could
107 provide a unique metric for measuring chemical weathering rates.

108 Other approaches for determining chemical weathering rates include contemporary estimates
109 from solute fluxes or long-term estimates based on changes in the elemental or mineralogical
110 distributions in the soil and some knowledge of the duration of weathering (*e.g.*, Brantley and
111 Lebedeva, 2011; Brantley et al., 2008; White, 2002; White et al., 2009; White et al., 2008). Such
112 approaches have provided robust estimates of chemical weathering rates if erosion rates are low
113 (*e.g.*, chronosequences). In addition to determining the depositional age, challenges for these
114 methods include reliable evaluation of the unweathered parent material in sedimentary
115 environments and uncertainties in the retention of immobile elements as the parent materials
116 weather. In a second approach, applicable to eroding surfaces, the denudation rate (sum of
117 erosion and weathering rate) at the surface is determined from *in situ* cosmogenic isotopes. By
118 assuming constant regolith thickness, the regolith production rate is then equal to the denudation
119 rate. In conjunction with immobile and mobile element concentrations, the weathering rate can
120 then be evaluated (Dixon and Riebe, 2014; Riebe et al., 2003).

121 Another approach to determining weathering rates relies on the redistribution of the
122 uranium-series (U-series) daughter isotopes over time. By solving a system of equations relating
123 the relative gain and loss of the ^{234}U and ^{230}Th daughters, the age of the weathering front can be
124 determined (Chabaux et al., 2013; Dosseto et al., 2012; Vigier et al., 2001; Vigier et al., 2006).
125 The depth of the weathering front divided by the U-series age gives the regolith production rate,
126 or the rate of downward migration of the weathering front. The U-series approach thus provides
127 a powerful tool for reconstructing chemical weathering rates. However, the method requires
128 numerous assumptions, including constant loss or gain of the daughter isotopes over time,
129 assumption that have been challenged by studies of chronosequences (Andersen et al., 2013;
130 Keech et al., 2013). Thus, new methods that complement existing approaches are necessary for
131 interrogating long-term weathering processes.

132 Due to the complex and time-dependent coupling between transport, sorption, and
133 weathering processes, simple mass balance descriptions of $^{10}\text{Be}/^9\text{Be}$ ratios and inventories cannot

capture the full complexity of multi-component weathering systems. One way to evaluate the inherent assumptions of parsimonious mass balance models and guide future data collection is to use multi-component reactive transport approaches to predict the evolution of ^{10}Be and $^{10}\text{Be}/^9\text{Be}$ in the aqueous, bulk solid and sorbed reservoirs over time and under a wide variety of environmental conditions, including variations in flow rates, weathering rates, erosion rates and chemical environments (*e.g.* soil water pH or variable sorption efficiency). Although such a modeling approach cannot capture all the processes that affect Be distributions (*i.e.*, illuviation, physical mixing in the mobile regolith, etc.), the rigorous mass balance and ability to perform sensitivity analyses on key parameters may complement extensive field measurement and/or laboratory experiments. Here we develop a set of differential mass balance equations for the $^{10}\text{Be}/^9\text{Be}$ composition of the bulk solid and sorbed reservoirs and evaluate them using a multi-component reactive transport model framework that includes the ability to impose variable water fluxes and soil production rates. The simulations represent granitic parent material and they reflect the deep (several meters) weathering zones typical for this lithology. We find that ^{10}Be inventories faithfully record the age of a weathering profile at neutral pH, but that if retention is incomplete, the ^9Be inventories cannot be used to reliably correct ^{10}Be inventories as incomplete retention affects each isotope to a different extent. Nevertheless, we find that $^{10}\text{Be}/^9\text{Be}$ directly reflects the chemical weathering rate. Thus, measurement of Be isotopes in the weathering zone provides a unique approach to separately constrain chemical weathering rates, which could be paired with *in situ* ^{10}Be denudation estimates to test long-standing assumptions about the processes that control the thickness of weathered material at the Earth's surface.

2. Methods

Although weathering processes are inherently three-dimensional, with water traveling vertically through the unsaturated zone and then laterally as groundwater while sediment is transported according to the divergence of the topographic gradients, one-dimensional descriptions still afford a useful perspective on the overall dynamics (Ferrier and Kirchner, 2008; Hilley and Porder, 2008; Lebedeva et al., 2010). A multi-component reactive transport model (RTM), CrunchFlow2011 (Steefel et al., 2015) was used to simulate weathering and sorption reactions and the attendant solute fluxes in one dimension over timescales typical of soil development (*i.e.* 0 to 600 kyr). The modeling framework was expanded to include ^9Be and ^{10}Be

isotopes, allowing for the isotope partitioning to be evaluated over time and space during the entire model duration (**Fig. 1**).

2.1 Description of isotope-enabled weathering simulations

The overall modeling and conceptual framework is shown in **Fig. 1**. Simulation conditions represent weathering of a granitic parent material and are similar to those presented in Maher and Chamberlain (2014), including experimentally determined rate constants and rate laws (**Table 1**). These simulations presented here consider infiltration fluxes from 0.1 to 1 m/yr and a range in Be mobility, resulting in 36 numerical realizations. The initial aqueous condition and upper boundary condition assume dilute rainwater in equilibrium with soil gas CO₂ at 0.005 bars and a constant water flux. For a full treatment of steady-state unsaturated flow, specification of a separate gas phase is required. This inclusion would not impact the mass balance or general relationships between flow and weathering processes (Maher, 2011; Maher et al., 2009); however, it would introduce additional uncertainty associated with variable amounts of CO₂(g) and the shape of the liquid saturation curve. Thus, as a simplification, profiles were assumed to be water saturated (*i.e.* liquid water saturation of 1.0). For the present study, we also assume no erosion in the simulations in order to evaluate “chronosequences” or soils of different ages. The weathering assemblage consists of quartz, K-feldspar and plagioclase (20% anorthite), with minor amounts of initial clay (**Table 1**). Two of the primary minerals, K-feldspar and plagioclase, are only allowed to dissolve, whereas the secondary phase, represented by a soluble precursor to kaolinite, halloysite, is allowed to dissolve and precipitate according to the saturation state of fluid ($\log(Q/K_{eq})$, where Q is the ion activity product and K_{eq} is the equilibrium constant. Quartz is assumed to be non-reactive.

In order to incorporate the Be isotope systematics, several modifications were made. First, the atmospheric fluxes of ⁹Be and ¹⁰Be into the weathering zone were set to constant values in all simulations by varying the concentration in the inlet fluid according to the prescribed infiltration flux. The ¹⁰Be flux (F^{10Be}) is 1.66×10^{-14} mol/m²/yr (1×10^6 atoms/cm²/yr), corresponding to mid-latitude values suggested in Willenbring and von Blanckenburg (2010b). The atmospheric ⁹Be flux (F^{9Be}) is 1×10^{-8} mol/m²/yr based on values measured in precipitation (Vesely et al., 2002). For comparison, the flux of weathering derived ⁹Be in the model simulations ranges from

ca. 1.6×10^{-6} to 1.5×10^{-5} mol/m²/yr. By extension, the $^{10}\text{Be}/^9\text{Be}$ of the input flux is held constant at 1.66×10^{-6} throughout the simulations.

Beryllium is an incompatible element during magmatic processes and thus concentrations in the primary silicate minerals vary according to the geochemical setting (Grew, 2002; Ryan and Langmuir, 1988), with highest concentrations in feldspars typically observed in sodic plagioclases within pegmatites (Grew, 2002). Despite the variability in Be concentrations, no systematic differences between plagioclase and K-feldspar are apparent (Grew, 2002). Thus, we assume average concentrations typical of the upper continental crust (von Blanckenburg et al., 2012) and equal concentrations in both primary silicates. The stoichiometries of K-feldspar and plagioclase were amended to include ^9Be as a trace element at a concentration of 2.5 ppm (von Blanckenburg et al., 2012), with no change in the equilibrium constants of the minerals. As a consequence, ^9Be in the model formulation is derived primarily from weathering of the primary minerals, whereas ^{10}Be is derived only from atmospheric deposition at a fixed flux (**Fig. 1**). Both ^9Be and ^{10}Be are assumed to be retained via sorption onto secondary mineral surfaces as described below. Because of the large variations in $^{10}\text{Be}/^9\text{Be}$, mass-dependent fractionation during sorption is considered to be negligible (von Blanckenburg et al., 2012). Decay of ^{10}Be in the aqueous and sorbed fractions is included in all simulations using a decay constant of $4.9975 \times 10^{-7} \text{ yr}^{-1}$ (Chmeleff et al., 2010; Korschinek et al., 2010).

Beryllium mobility in the weathering zone is governed by aqueous speciation and sorption onto mineral surfaces (Aldahan et al., 1999; Vesely et al., 2002; You et al., 1989). Beryllium is differentiated from other Group IIA elements in that the d-orbital is unoccupied and it has a small ionic radius such that in aqueous solutions the high charge to radius results in strong interactions with water molecules (Hinz et al., 1986). To account for this behavior, the predominant aqueous Be species as a function of pH were added to the secondary species list: $^9\text{Be}^{2+}$, $^{10}\text{Be}^{2+}$, $^9\text{Be}(\text{OH})_2(\text{aq})$, $^{10}\text{Be}(\text{OH})_2(\text{aq})$, $^9\text{Be}_2\text{OH}^{3+}$, $^{10}\text{Be}_2\text{OH}^{3+}$. The formation and importance of an additional aqueous species, $\text{Be}(\text{OH})^+$, is debated. Given the likely minor importance of $\text{Be}(\text{OH})^+$ at low pH relative to Be^{2+} and $\text{Be}(\text{OH})_2(\text{aq})$ species (Hinz et al., 1986) and the large variation in equilibrium constants, it was not included in the calculations. Organic acids can also form strong Be complexes that can reduce the sorption depending on the pH and substrate (Takahashi et al., 1999). Although organic acids may be present in the weathering zone, particularly at shallow depths, their concentrations are generally uncertain and can vary

widely among weathering environments (Jones, 1998; Lawrence et al., 2014). In addition, there are very few equilibrium constants for organo-beryllium complexes. Thus, to avoid introducing additional parameters, complexation of Be by organic acids was not included in the present simulations. Instead, the effect of poor retention, either due to organic acids, competition for surface sites, etc. is addressed in a general way by varying the extent of sorption from nearly complete retention to almost no retention.

Biological cycling may amplify differences in the redistribution of the Be isotopes, given their different sources. As our simulations extend out to 600 kyr and our focus is on the mass balance within the weathering zone, not the ultimate drivers of weathering, we assume that biological cycling of Be does not impact the fundamental mass balance or general relationships between ^9Be and ^{10}Be over such long timescales. In addition, average ^9Be concentrations in plants range from 10-100 ppb compared to 2500 ppb in soil/rock (Vesely et al., 2002) suggesting the ^9Be budget is likely dominated by rock-derived Be. Nevertheless, additional work on the role of biological cycling in controlling Be distributions is needed to fully address this assumption.

2.3 Solubility of Be as a function of pH

Naturally occurring Be-hydroxides are rare and are mostly found associated with weathering of Be-rich deposits, such as pegmatites (Ehlmann and Mitchell, 1970). Nevertheless, the solubility of secondary Be phases is an important consideration in the design of sorption experiments. Depending on the aqueous Be concentration, Be solubility drops appreciably as pH increases above circumneutral, resulting in the formation of amorphous $\text{Be}(\text{OH})_2(\text{s})$ according to the reaction:

At pH 7, Be concentrations above 1.5×10^{-7} mol/L (1.4 ppb) are predicted to result in formation of pure Be solids (based on a $\log(K_{eq})$ of 7.19 at 25°C and assuming $[\text{Be}^{2+}] \approx \log(K_{eq}) - 2\text{pH}$). For sorption experiments that exceed the theoretical solubility of $\text{Be}(\text{OH})_2(\text{s})$, precipitation may occur (as noted by Boschi and Willenbring (2016)), and thus we do not consider experimental data above this limit in the development of the surface complexation model described below. At

the concentrations predicted in the modeling ($<10^{-9}$ mol/L) the $\text{Be}(\text{OH})_2$ was undersaturated and thus it was not included in the simulations.

2.2 Development of surface complexation model for Be

Relatively few studies have focused on Be sorption under environmentally relevant conditions due to the difficulty of measuring low concentrations of aqueous Be. Most studies have also used a traditional partition coefficient (K_d) approach for describing the sorption of Be onto mineral surfaces. The K_d value for Be sorption is commonly defined according to the amount of Be sorbed per mass of solids, $[\text{Be}]_{\text{reac}}$, relative to the dissolved concentration, $[\text{Be}]_{\text{diss}}$ (Aldahan et al., 1999; Brown et al., 1992; You et al., 1989):

$$K_d (\text{L} / \text{kg}) = \frac{[\text{Be}]_{\text{reac}}}{[\text{Be}]_{\text{diss}}} . \quad (3)$$

This description is difficult to apply in soil zones because the K_d value is known to be pH-dependent (e.g., 10^3 ml/g at pH 4 to 5, 10^4 ml/g at pH 5 and 6 and 10^5 ml/g above pH 6 (Aldahan et al., 1999; You et al., 1989). In addition, Be sorption in soils should be linked to the amount of available mineral surface area (Dzombak and Morel, 1990). Thus, as secondary minerals accumulate (or dissolve) in the soil profile over time, the K_d value is expected to be time dependent.

Because the extent of Be sorption in the weathering zone is a function of pH, time, and the nature of the available mineral surfaces, an alternative approach for modeling Be retention on mineral surfaces is to use a surface complexation model (Davis et al., 1998; Hayes et al., 1991; Sverjensky and Sahai, 1996). There are multiple models of varying complexity that can be used to describe inner-sphere surface complexation; these differ from those used to describe cation exchange or outer-sphere surface complexation (Goldberg, 1992). The disadvantage of surface complexation models is that they require considerably more data to parameterize, including knowledge of the distribution of surface sites, the equilibrium constants describing protonation and de-protonation of the surface sites, and the speciation of the surface complexes. Nevertheless, simplified surface complexation approaches have been successfully applied to model the retention of metals and actinides on complex porous media (Davis et al., 1998; Davis et al., 2004; Mayer et al., 2002).

Here, a simplified surface complexation description was developed using Be sorption data from the literature and the existing surface complexation model framework included in CrunchFlow (Steefel et al., 2015). Although numerous studies have evaluated Be sorption, only a few studies conducted experiments at low concentrations suitable for modeling natural systems, and in some cases the concentrations exceeded the solubility of Be-hydroxides at higher pHs (section 2.3). You et al. (1989) used a ^7Be tracer to quantify K_d values as a function of pH for a range of different sediments at total Be concentrations of 1×10^{-9} mol/L. Brown et al. (1992) and Wittmann et al. (2015) collected dissolved and particulate riverine material to calculate *in situ* K_d values for bulk suspended sediment. Aldahan et al. (1999) also conducted sorption experiments on mineral separates, although only the data at or below pH 6 is likely to be unaffected by $\text{Be}(\text{OH})_2(\text{s})$ precipitation. Several studies also used high initial dissolved Be concentrations (~ 0.1 to 0.5 mmol/L) (Aldahan et al., 1999; Boschi and Willenbring, 2016) that exceed those of natural waters by up to several orders of magnitude (natural waters typically range from 0.3 to 50 nmol/L (Vesely et al., 2002)). Thus, in developing the surface complexation model (**Fig. 2**) we consider only K_d values measured in natural systems or at concentrations representative of natural waters. Ideally, a surface complexation model for each mineral would be developed and incorporated into the reactive transport model. Unfortunately, several key substrates which are known to strongly sorb trace metals are not represented in the existing data, namely Fe- and Mn-(oxy)hydroxides and double-layer clays such as smectite. In the experimental sediments, these materials were likely mixed with less reactive and lower surface area substrates such as quartz and feldspar, suggesting that the actual K_d values for the more reactive substrates may be higher than indicated by the existing data. This inference is supported by the relatively low K_d values for albite and biotite (Aldahan et al. 1999). In addition, Fe and Mn-oxides may undergo reductive dissolution in soils or sediments which would release any sorbed or occluded Be, altering the K_d values for bulk soil or sediment. The above discussion outlines the need for more data on both the aqueous speciation and sorption of Be under natural conditions.

Given the broad range of values and limited data, Be sorption is treated as a variable in the simulations. This enables us to further evaluate the effect of poor retention on the $^{10}\text{Be}/^9\text{Be}$ systematics. In order to capture the general characteristics of the observed range of K_d values, we assume that primary minerals are weathering to secondary minerals, including clays and Fe-(oxy)hydroxides. As we do not specifically model Fe oxidation and reduction, we assume that

the secondary mineral assemblage is correlated with the halloysite abundance as commonly observed during weathering (Brantley et al., 2008; White et al., 2008). Thus, halloysite (kaolinite) is used as a proxy for the amount of substrate available to sorb Be. Furthermore, we assume that the secondary mineral assemblage has sorption characteristics similar to that of Fe-oxides, with deprotonation, protonation and Be surface complex values estimated from Dzombak and Morel (1990); (**Table 2**). This assumption is further justified because Fe-oxides often dominate the reactive surface area of soils and sediments (Cornell and Schwertmann, 2003). **Table 2** summarizes the reactions and assumed values for the equilibrium constants describing protonation and deprotonation of the secondary minerals, and the sorption of Be. Reactions 1 through 3 in **Table 2** are linked to the amount of kaolinite produced, so that as kaolinite volume fraction and surface area increases, the amount of sorbed Be increases as well. Because ^9Be is mostly liberated at the weathering front, whereas ^{10}Be is deposited at the surface, the isotopes are exposed to different amounts of surface area, this parameterization allows us to test the extent to which ^9Be loss can be used to evaluate ^{10}Be loss.

The results of applying the assumed values above are shown in **Fig. 2**. The model curves correspond to a reaction path model with particle density of 0.2 g/L from You et al. (1989). In general, the assumed parameters capture the general pH dependence of the existing data. In order to bracket the total range of observed K_d values, the site density was varied from the maximum of $1.1 \times 10^{-7} \text{ mol/m}^2$ associated with Fe-oxides to a lower-end value of $1 \times 10^{-9} \text{ mol/m}^2$. As noted above, this may be due to variable experimental conditions and the mixture of multiple substrates within the experiments. As a reference model, a site density of $5 \times 10^{-8} \text{ mol/m}^2$ was selected to represent a reasonable value for retention on the secondary phases. For comparison to previous studies, bulk K_d values calculated from the model will be shown in subsequent sections.

2.4 Description of simulations

In order to evaluate the coupled evolution of ^{10}Be and ^9Be across a range of conditions (namely varying weathering rate and varying retentivity), we consider “chronosequence” evolution, where erosion rates are negligible and thus the weathering front deepens over time according to the weathering rates. These conditions might represent sedimentary deposits, including marine terrace deposits (Lawrence et al., 2014; Maher et al., 2009; White et al., 2009; White et al., 2008) fluvial deposits (White et al., 1996), various glacial deposits (Blum and Erel,

1995), and abandoned alluvial fan surfaces (Chabaux et al., 2012; Maher et al., 2007; Oster et al., 2012). The model domain consists of 500 nodes of 0.05 m for a total depth of 25 meters. Four different infiltration rates were considered (0.1, 0.3, 0.5 and 1 mm/yr) and the simulations were run forward to 600 kyr with output tabulated at 0, 5, 25, 50, 100, 200, 300, 400, 500 and 600 kyr. Each set of infiltration fluxes was run with nine different site densities (corresponding to those in **Fig. 2**) resulting in 36 numerical simulations. A site density of 5×10^{-8} mol/m² at each flow rate is used as a reference simulation corresponding to full Be retention. For each simulation the sorbed ¹⁰Be/⁹Be at each depth was compared to the dissolved and in all cases the model reflected equilibrium partitioning suggesting that mass balance was always honored.

2.5 Calculations

The reactive transport model tracks mineral volumes ($V_{m,j}$) [m³/m³ bulk porous media], bulk surface areas (A_j) [m²/m³], porosity (ϕ) [m²/m³], and aqueous (C_i) [mol/L] and sorbed concentrations ($S_{j,i}$) [mol/m³] over space and time (where index j refers to the solid phase and i refers to the chemical component). A table of parameter and definitions is provided in **Table 3**. Collectively, these parameters define the physical and chemical aspects of the system of interest. However, these values must be converted into measures relevant to ¹⁰Be/⁹Be systematics. The following conversions are used.

Aqueous fluxes are calculated by multiplying the concentrations times the infiltration flux. The instantaneous solute-based net chemical weathering rate (W) is calculated by summing the concentrations of K, Na, Ca, Si and Al (there is no Mg-bearing mineral in the simulations). The W values are comparable to those obtained from the time-integrated mineral depletion, particularly in latter stages of the simulation. The range of weathering rates simulated spans from ca. 2.5 to 25 g/m²/yr.

Sorption is output from the model as the amount of Be sorbed per volume of bulk porous media. To calculate the inventory (I_i), each value is multiplied by the depth interval for of the model node (0.05 m) and then summed accordingly. The corresponding model K_d value is calculated two ways (using either the kaolinite or the total volume of solids) by converting the sorbed Be concentration into a mass concentration using the volume fraction of solid and the density (2.6 g/cm³).

The solid inventories and aqueous profiles and fluxes are evaluated over two depths. The first approach is to evaluate the solid inventory over the depth of the weathering front, defined as the depth at which the porosity is more than 0.1% different from the protolith (*i.e.*, $\phi > 15.1\%$). This depth generally corresponds to the location of the plagioclase weathering front as it is the first mineral to dissolve at the weathering front. The second approach is to tabulate the solid inventories over the entire 25 m domain. Both approaches yield identical results and thus only the first approach is presented in supplemental table S1. In simulations where the weathering front exceeded the 25 m model domain (usually at high infiltration flux), only the time points when the values were within the domain are used in calculations, although all are tabulated in the results. For both depth intervals, the depletion of primary minerals and accumulation of secondary minerals at each time output is calculated using the difference between the current and initial volume fractions. The depletion of primary minerals indicates the gross time-integrated weathering flux. The net weathering flux, corresponding to the aqueous solute flux, is calculated using the solid phase concentrations relative to the parent material.

3. Results

The evolution of the solid phase profiles over depth and time control both the sorbed and aqueous Be profiles. Conceptually, increases in the flow rate increase the net weathering flux, altering the solid profiles that determine the Be distributions. The Be distributions are further modified by variations in Be retention. Thus, in order to illustrate the importance of the water flux, we show two representative simulations: “arid” conditions of 0.1 m/yr and more “humid” conditions of 0.5 m/yr. Similarly, to illustrate the effect of variable retention on the Be profiles and inventories we show two site densities: the reference value of 5×10^{-8} mol/m² and a lower-end value of 5×10^{-9} mol/m². Supplementary Table 1 contains the results of the numerical simulations presented below.

3.1 Results for chronsequence simulations of Be accumulation during weathering

The temporal evolution of the weathering zones between relatively arid ($q = 0.1$ m/yr) to relatively humid ($q = 0.5$ m/yr) is shown in **Fig. 3**. (Note the increase in the profile age interval at >100 kyr). In the arid case, the mineral depletion fronts propagate downward with time, while the secondary mineral profile reflects the sum of the plagioclase and K-feldspar profiles resulting in a sharp peak corresponding to K-feldspar dissolution and a more diffusive peak corresponding

to the plagioclase dissolution front. The kinetic rate constants and surface areas of the two minerals are broadly similar (Table 2), hence the deeper reaction front for plagioclase is partly attributed to the much greater solubility of plagioclase relative to K-feldspar (Maher et al., 2009; White et al., 2008). The gradient in the mineral depletion profiles is also controlled by a complex set of processes. The base of the weathering front is controlled by the approach to equilibrium: as the fluid approaches equilibrium along the flow the driving force for dissolution is diminished and the dissolution rates approach zero (*e.g.*, **Fig. 3a**). A more soluble phase, like plagioclase, requires a longer fluid residence time to reach equilibrium (*e.g.*, **Fig. 3b**). At the top of the profiles, the mineral volume and hence surface area approaches zero, decreasing the dissolution rate (Lebedeva et al., 2010; Maher, 2010). As the flow rate increases the approach to equilibrium is extended, resulting in a steeper gradient for K-feldspar (*e.g.*, **Fig. 3e**) or a nearly uniform decrease in mineral volumes with depth for plagioclase (*e.g.*, **Fig. 3f**). Thus, in the humid case, the increase in water flux results in deeper weathering profiles and faster weathering advance rates which results in more kaolinite accumulation at depth relative to the arid case. The difference in weathering rates between the two cases is approximately equal to the difference between the flow rates, or a factor of 5.

The effect of linking the amount of surface area available for sorption to the accumulation of secondary minerals results in variable K_d values as a function of time and depth (**Fig. 4**). Here, using the sorbed concentrations and mineral volumes, the K_d values associated with ^9Be sorption on the bulk sediment are calculated from the sorbed and dissolved ^9Be concentrations for the arid case. Like the reactive Be content, the K_d values generally track the kaolinite profile. The effect of decreasing the site density to $5 \times 10^{-9} \text{ mol/m}^2$ (**Fig. 4b**) leads not only to much lower K_d values but also to greater spread as a function of depth. This suggests that as the site density (or retention of Be) decreases, the difference between the amount of sorption at the weathering front (where ^9Be is released) compared to the top of profile where ^{10}Be is deposited, could be substantially different. The consequences for correcting using ^9Be to correct for ^{10}Be loss will be explored in a subsequent section.

As summarized in **Fig. 2**, the range of K_d values at a pH of 6 to 7 are between about 3×10^4 and $1 \times 10^6 \text{ ml/g}$. The K_d values in the model for the reference case (**Fig. 4a**) range from initial values of $1 \times 10^3 \text{ ml/g}$ to a maximum of $2.6 \times 10^5 \text{ ml/g}$ at shallow depths/maximum clay accumulation, well within the observed range. When the site density is decreased by an order of

magnitude from the reference case, the K_d values decrease to initial values of 15 ml/g and a maximum of 1.2×10^4 ml/g, below the values reported in the literature for Be retention on clays (**Fig. 4b**). Thus, the reference model (**Fig. 4a**) is a reasonable reflection of expected weathering zone values. The selection of the lower site density as the low retention case also results in values that are consistent with the range of values observed in the compilation of **Fig. 2**.

The importance of the water flux can also be seen in the aqueous profiles (**Fig. 5**). In the arid case, $\text{SiO}_2(\text{aq})$ concentrations, a useful proxy for weathering rates in the model, reach a nearly constant value at depth (**Fig. 5a**) whereas in the humid case the concentrations at depth vary over time (**Fig. 5e**). The pH evolution also varies due to the different flow rates and extents of water rock interaction, with higher pH values associated with the arid simulation as the reactions reach equilibrium (generating more alkalinity) over a shorter depth (see **Fig. 3**). In the humid simulations, the higher flux of relatively low-pH CO_2 -rich fluid prolongs the approach to equilibrium resulting in lower pH values in the upper meters that may reduce Be retention. The dissolved ^9Be ($[^9\text{Be}]_{\text{diss}}$) profiles, corresponding to the reference values for retention (e.g., a site density of $5 \times 10^{-8} \text{ mol/m}^2$) are also markedly different. In contrast to $\text{SiO}_2(\text{aq})$, ^9Be dissolved concentrations are at a minimum where kaolinite volume fractions are at a maximum and then increase again at the weathering front, emphasizing the coupling between the long-term solid evolution and the spatial variations in accumulation of Be under different climates. The $[^{10}\text{Be}]_{\text{diss}}$ profiles are consistent with nearly complete retention in the upper portions of the weathering zone. The difference between the $[^{10}\text{Be}]_{\text{diss}}$ and $[^9\text{Be}]_{\text{diss}}$ distributions in the arid and humid climate cases can be attributed the effect of the water flux and the retardation factor, R_f , which can be defined in terms of the K_d value:

$$R_f = 1 + \frac{K_d \rho_b}{\phi} \quad (4)$$

where ρ_b is the bulk density and ϕ is the porosity. The difference between the depth of the sorption fronts is thus proportional to $q \times R_f$. As a result, the depth of the ^{10}Be sorption front in the arid simulations is about 0.5 m and very little dissolved Be has migrated downward compared to the humid case where the sorption front has reached 2.5 m. As a general rule of thumb, for the same retentivity the depth of the ^{10}Be sorption is proportional to the relative difference between the flow rates.

The corresponding sorbed or “*reac*” profiles are shown in **Fig. 6** for the reference simulation. The $[^9\text{Be}]_{\text{reac}}$ profiles are a reflection of the kaolinite profiles in **Fig. 3**. In contrast, the $[^{10}\text{Be}]_{\text{reac}}$ profiles are closer in appearance to the $[^{10}\text{Be}]_{\text{diss}}$ profiles but show deeper penetration of ^{10}Be , consistent with a strongly sorbing element (the integrated fluxes of ^{10}Be to the profiles are identical). Under more humid conditions, the ^{10}Be propagates further into the weathering zone. When the $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ profiles are compared, both show a decrease with depth as expected.

3.2 Age dating of weathering surfaces using $[^{10}\text{Be}]_{\text{reac}}$

Although ^{10}Be has been used to date alluvial surfaces (Duhnforth et al., 2012; Egli et al., 2010) and determine soil residence times (Bacon et al., 2012; Barg et al., 1997; Brown et al., 1992; Pavich et al., 1986), poor Be retention is a key uncertainty. **Fig. 7** illustrates the potential for full retention of the $[^{10}\text{Be}]_{\text{reac}}$ across the broad range of climate and weathering rates accessed in the simulations. Although the $[^{10}\text{Be}]_{\text{reac}}$ depth profiles become deeper and more diffuse with increasing flow rate, the computed inventories are in perfect agreement with the closed-system estimate generated by multiplying the flux ($F^{10\text{Be}}$) by the duration of the simulation and accounting for radioactive decay (**Fig. 7a**). In contrast, at low retentivity (as in **Figs. 6b** and **7b**), the inventories no longer reflect the closed-system values required for geochronology. Interestingly, the open system behavior is the most severe in the arid case because of the lower amount of kaolinite accumulation. The loss in accuracy on the ages under these conditions would be between 51% in the arid case (cumulative ^{10}Be loss fraction of 50%) and 35% for the most humid case (cumulative ^{10}Be loss fraction of 34%).

3.3 Relationship between $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ and chemical weathering rates

The depth profiles of reactive, dissolved, and primary Be are complex given the dynamics of the conversion to secondary minerals coupled with the flow and sorption. The inventories show a more straightforward behavior when evaluated at different flow rates (corresponding to different weathering rates) at the same model time (250 kyr) (**Fig. 8a**). The chemical weathering rate (*i.e.*, the dissolved mass lost from the weathering zone) in the arid simulation is $2.5 \text{ g/m}^2/\text{yr}$ compared to $25 \text{ g/m}^2/\text{yr}$ at the highest flow rate of 1 m/yr . The $(I_{10\text{Be}}/I_{9\text{Be}})_{\text{reac}}$, as well as the $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$, profiles, reflect the greater input of ^9Be to the reactive pool as weathering rates increase. Thus, at high retention the $(I_{10\text{Be}}/I_{9\text{Be}})_{\text{reac}}$ is inversely proportional to the chemical weathering rate.

Because of the different controls on the partitioning of the two Be isotopes over space and time, it is the inventory and not the gradient of the $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ that records the weathering rate, even at low retention.

At low retention, a systematic relationship between the inventory and the weathering rate is still apparent, although the difference between the high- and low-retention cases is greatest at the high flow rates (**Fig. 8b**). The depth profiles at the low retention case also look very similar across the four flow rates because at lower retention the spatial dependence that arises from the two different input sources of Be is less apparent. Although we considered only one lithology, our results emphasize the importance of both processes at the weathering front and the rate of secondary mineral accumulation. At low flow rates, where the weathering front is thin and sharp, we see greater ^{10}Be loss compared to the higher flow rates characterized by diffuse and more evolved weathering fronts. The opposite is true for ^9Be , where loss fractions increase slightly with increasing weathering rate, although overall ^9Be loss fractions are less sensitive to the weathering rate. Basaltic parent rock that develops thinner weathering zones (Bazilevskaya et al., 2015) would thus be an important target for future modeling studies combined with field measurements. Given the challenges modeling mafic bedrock transformations, a full treatment of this lithology is beyond the scope of the current paper.

3.4 Beryllium-9 loss fractions as a correction for poor retention of ^{10}Be

If ^9Be loss fractions could be used to correct for incomplete retention of ^{10}Be this would provide a useful approach for using the ^{10}Be inventories to date surfaces and using the $(I_{^{10}\text{Be}}/I_{^9\text{Be}})_{\text{reac}}$ to calculate weathering rates. The temporal changes in the inventories of both isotopes are shown in **Figs. 9a and b** for the arid end-member case and are tabulated according to the specified time outputs. Both inventories show the expected time-dependent increases. We also show the agreement between the inventory as a function of age and the closed-system (equation 2) and the $I_{^{10}\text{Be},\text{reac}}$ of the model. For $^{10}\text{Be}_{\text{reac}}$, the non-linear increase in the inventory at lower retentions is due to greater loss rates at early times when secondary mineral surfaces are less abundant, particularly at the top of the profile. **Figure 9c** shows the corresponding $(I_{^{10}\text{Be}}/I_{^9\text{Be}})_{\text{reac}}$ evolution over time. For high retention cases, the $(I_{^{10}\text{Be}}/I_{^9\text{Be}})_{\text{reac}}$ decreases linearly with time as ^9Be is added to the inventory slightly faster than ^{10}Be . As the retentivity decreases, the pattern switches and $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ increases with time because the loss of ^9Be is maximized

at the weathering front where secondary mineral volumes are low. This can also be seen by comparing the two loss fractions. At high retention, some ^9Be is still lost from the weathering zone at the weathering front, whereas all ^{10}Be is retained. As the retentivity of Be decreases, the difference becomes more pronounced at moderate Be loss. The shape of the curves also reflects the temporal evolution of the loss fractions, where the loss fraction ^9Be increases more rapidly with increasing time. In contrast, the loss fraction of ^{10}Be reaches a maximum early on and then decreases with time, resulting in the humped patterns shown in **Figure 9d**. In summary, the model results suggest that using ^9Be loss fractions to correct ^{10}Be inventories may result in over correction. Nevertheless, such a correction may provide a useful maximum age estimate.

4. Discussion

4.1 Beryllium inventories as an age dating tool

The application of ^{10}Be dating and $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ offers a unique approach to determine both the surface ages and the chemical weathering rates. Although the numerical simulations we present are highly idealized, the results suggest that given sufficient Be retention, the $I_{10\text{Be},\text{reac}}$ records the time since the surface started to accumulate ^{10}Be . For cases in which ^{10}Be retention is incomplete we have evaluated whether the concomitant decrease in $I_{9\text{Be},\text{reac}}$ can be used to (1) identify the amount of ^{10}Be that might have been lost from the profile, (2) correct for ^{10}Be loss, and (3) provide confidence intervals for the age estimate that encompass the unknown loss of ^{10}Be . The error on the age introduced purely by Be loss, computed from the inventories, is shown in **Fig. 10a**. For both the arid and the humid cases, at high to moderate retention the error on the age is minimal. At lower retentivity, the bias on the age is larger in the more arid case because of lower rates of secondary mineral accumulation. There is also a maxima in the uncertainties at approximately 100 kyr, at which point the error values decrease as more secondary minerals are added at depth. Thus the effect of poor ^{10}Be retention on the age appears to be highly time dependent.

In **Fig. 10b** we evaluate the use of the loss fraction determined from the ^9Be inventories to correct for ^{10}Be loss. Our simulations suggest that because ^{10}Be enters the weathering zone from the surface, whereas ^9Be is released predominantly at the active weathering front, using ^9Be loss to correct for ^{10}Be loss may overestimate the ^{10}Be loss, and thus provides a maximum uncertainty on the age (**Fig. 10b**). For example, at a slightly decreased retention ($2.5 \times 10^{-8} \text{ mol/m}^2$), the ^9Be

loss fraction is on the order of 10-14%, whereas the corresponding ^{10}Be loss fraction is ca. 0.1%, resulting in an over correction of the ^{10}Be age (**Fig. 10a**). Correcting for ^{10}Be loss using each depth point in the profile before summing to obtain the inventory magnifies the error substantially because ^9Be loss is most pronounced at the surface as the shallow ^9Be is released and flushed downward early in the model, whereas ^{10}Be is retained. For comparison, correcting the depth profile for the full retention case at the low flow rate ($q = 0.1 \text{ m/yr}$) results in an over-estimation of the age by 24%, compared to a negligible over-correction when the inventory is used. We note that for the young ages commonly of interest for dating of late Quaternary geomorphic features (*i.e.*, $t < 20 \text{ ka}$), the $I_{9\text{Be},\text{reac}}$ correction for loss introduces a minor error only. Thus, dating is facilitated if the exposed features are either young, or else show high retention potential.

4.2 Evaluating $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ as weathering rate meter using differential mass balance approaches

The numerical experiments presented here suggest that the $(I_{10\text{Be}}/I_{9\text{Be}})_{\text{reac}}$ is inversely proportional to the time-integrated weathering rate, as long as most of the Be is retained (**Fig. 8**). To further evaluate the potential utility of $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$, we compare the results of the reactive transport model to equations for the differential mass balance in order to provide an analytical framework that directly relates $(I_{10\text{Be}}/I_{9\text{Be}})_{\text{reac}}$ to chemical weathering rates. Although the present work is focused on chronosequences, we include the effect of erosion for completeness. The $(I_{10\text{Be}}/I_{9\text{Be}})_{\text{reac}}$ records the weathering rate because the ^{10}Be accumulation is a function of the depositional flux ($F^{10\text{Be}}$) and the removal via erosion (E), such that by mass balance the inventory records either absolute time (t) or soil residence time (τ) at high E , whereas the inventory of ^9Be at any given time indirectly reflects the production of regolith mass. Thus, we observe the inverse relationship between $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ and weathering rate in the model simulations (**Fig. 8**). This can be expressed mathematically by starting with the differential mass balance for the inventory of primary mineral ($M_{\text{prim}} [\text{g/m}^2]$) as a function of the erosion rate ($E [\text{g/m}^2/\text{yr}]$), the supply rate of unweathered material into the weathering zone through regolith production ($RP_{\text{prim}} [\text{g/m}^2/\text{yr}]$), and the removal by chemical weathering described as the product of the dissolution rate constant ($k_w [\text{yr}^{-1}]$) and the mass of primary minerals in the weathering zone

(Bouchez et al., 2013; Brantley and Lebedeva, 2011; Brantley et al., 2008; Ferrier and Kirchner, 2008; Lebedeva et al., 2010):

$$\frac{dM_{prim}}{dt} = RP_{prim} - k_w M_{prim} - E_{prim} \quad (5)$$

Following Bouchez et al. (2013), we use the subscripts *prim* and *sec* to refer to primary and secondary minerals, respectively, whereas *tot* refers to both combined. An element retained on or within a secondary mineral is specified by the subscript *reac*. The rate of secondary mineral accumulation in the weathering zone is similarly defined by the fraction of primary mineral converted into secondary minerals (f_{sec}):

$$\frac{dM_{sec}}{dt} = f_{sec} k_w M_{prim} - E_{sec} \quad (6)$$

We assume in this analysis that secondary minerals represent the stable end-product of weathering and remain in the weathering zone once they have formed such that the weathering rate (W [g/m²/yr]), the dissolved mass loss from the weathering zone, is the difference between the primary mineral dissolution rate and the rate of secondary mineral accumulation. Combining the expressions, we obtain an equation for the total mass of primary and secondary mineral in the weathering zone:

$$\frac{dM_{tot}}{dt} = RP_{prim} - (1 - f_{sec}) k_w M_{prim} - E_{tot} \quad (7)$$

$$\frac{dM_{tot}}{dt} = RP_{prim} - (1 - f_{sec}) k_w X_{prim} M_{tot} - E_{tot} \quad (7a)$$

where the right-hand side of equations (7) and (7a) are equivalent to $RP_{prim} - W - E$. For the chronosequence cases considered here, where $E = 0$, the equation is:

$$\frac{dM_{tot}}{dt} = RP_{prim} - (1 - f_{sec}) k_w X_{prim} M_{tot} \quad (8)$$

where X_{prim} is concentration of the primary mineral in the weathering zone. The net chemical weathering rate, W , is thus equal to $(1 - f_{sec}) k_w X_{prim} M_{tot}$. For the simulations presented above, W calculated from the bulk elemental concentrations is linearly proportional to the primary mineral dissolution rate, suggesting a constant value of $(1 - f_{sec})$ of 0.47 for these simulations (**Fig. 11a**).

This value is consistent with the stoichiometry of the weathering reactions contained in the model, where 1 mol of plagioclase and 1 mol of K-feldspar combine to form 1.1 mol of kaolinite. When converted into mass fractions, this stoichiometry results in a similar value of $1-f_{sec}$ of 0.477.

Next we combine this physical framework with the individual concentrations of $[^9\text{Be}]_{\text{reac}}$ and $[^{10}\text{Be}]_{\text{reac}}$ in the regolith $[\text{mol Be/g}_{\text{tot-regolith}}]$. Thus, equation 7 serves as the basis for the differential mass balance involving the individual isotopes. For the primary mineral-bound ^9Be ($[^9\text{Be}]_{\text{min}}$), the concentration in the regolith is the balance among the regolith production, the amount dissolved from parent material ($[\text{Be}]_{\text{parent}}$) that has entered the weathering zone, and the amount removed by erosion:

$$M_{\text{tot}} \frac{d[^9\text{Be}]_{\text{min}}}{dt} + [^9\text{Be}]_{\text{min}} \frac{dM_{\text{tot}}}{dt} = RP_{\text{prim}} [^9\text{Be}]_{\text{parent}} - k_w X_{\text{prim}} M_{\text{tot}} [^9\text{Be}]_{\text{parent}} - E_{\text{tot}} [^9\text{Be}]_{\text{min}} \quad (9)$$

and for the $[^9\text{Be}]_{\text{reac}}$ associated with secondary minerals it is the difference between the gross weathering flux and the erosion rate:

$$M_{\text{tot}} \frac{d[^9\text{Be}]_{\text{reac}}}{dt} + [^9\text{Be}]_{\text{reac}} \frac{dM_{\text{tot}}}{dt} = f_{\text{sec}}^{9\text{Be}} k_w X_{\text{prim}} M_{\text{tot}} [^9\text{Be}]_{\text{parent}} - E_{\text{tot}} [^9\text{Be}]_{\text{reac}} \quad (10)$$

where $f_{\text{sec}}^{9\text{Be}}$ is the fraction of weathered ^9Be retained in secondary minerals, with a value of one representing complete retention (note that this fraction differs from the flux fraction, f_{reac}^9 , used in von Blanckenburg et al. (2012) as discussed below). In our simulations with high retention rates this value is very close to 1. Thus, the first term on the right-hand side of equation 10 represents the accumulation rate of secondary ^9Be from weathering. As shown in **Fig. 9b**, the $[^9\text{Be}]_{\text{reac}}$ inventory increases at a constant rate depending on the efficiency of Be retention.

The differential mass balance equation for ^{10}Be is similar, but contains a term for production, $F^{10\text{Be}}$ $[\text{mol/m}^2/\text{yr}]$, radioactive decay, and a term for loss describing any ^{10}Be that is not retained in the secondary minerals or is subsequently leached (L^{10} $[\text{yr}^{-1}]$). For comparison, the loss of ^9Be is incorporated into equation (10) by considering $1 - f_{\text{sec}}^{9\text{Be}}$, which describes the fraction of ^9Be

dissolved from primary minerals that is not retained in the regolith. The resulting equation is:

$$M_{tot} \frac{d[^{10}\text{Be}]_{\text{reac}}}{dt} + [^{10}\text{Be}]_{\text{reac}} \frac{dM_{tot}}{dt} = F^{10\text{Be}} - E_{tot} [^{10}\text{Be}]_{\text{reac}} - L^{10} M_{tot} [^{10}\text{Be}]_{\text{reac}} - / M_{tot} [^{10}\text{Be}]_{\text{reac}} \quad (11)$$

The two differential mass balance equations (*i.e.*, equations 10 and 11) are combined by applying the quotient rule to the isotope mass balances above (*e.g.*, Fantle et al., 2010; Maher et al., 2003). Because we are assuming depth integrated profiles, we use the ratios of the inventories, $(I_{10\text{Be}}/I_{9\text{Be}})_{\text{reac}}$, to represent r_{reac} . Accordingly, equation (10) is multiplied by r_{reac} :

$$r_{\text{reac}} M_{tot} \frac{d[{}^9\text{Be}]_{\text{reac}}}{dt} = r_{\text{reac}} f_{\text{sec}}^{9\text{Be}} k_w X_{\text{prim}} M_{tot} [{}^9\text{Be}]_{\text{par}} - r_{\text{reac}} E_{tot} [{}^9\text{Be}]_{\text{reac}} - r_{\text{reac}} [{}^9\text{Be}]_{\text{reac}} \frac{dM_{tot}}{dt} \quad (12)$$

Then equation 12 is subtracted from equation 11 to yield the relationship between the rate of change of r_{reac} and the individual fluxes. Rearranging, we obtain a simplified equation for r_{reac} :

$$\frac{dr_{\text{reac}}}{dt} = \frac{F^{10\text{Be}}}{M_{tot}(t) [{}^9\text{Be}]_{\text{reac}}} - \frac{r_{\text{reac}} f_{\text{sec}}^{9\text{Be}} k_w X_{\text{prim}}(t) [{}^9\text{Be}]_{\text{parent}}}{[{}^9\text{Be}]_{\text{reac}}} - L^{10} r_{\text{reac}} - / r_{\text{reac}} \quad (13)$$

where we note that the M_{tot} and X_{prim} are also both a function of time. When the isotope ratio equation is generated, the terms containing E and dM_{tot}/dt cancel out. Conceptually, erosion removes material at the isotopic composition of the weathering zone and thus should not impact the isotope ratio directly. However, E may play a role in moderating M_{tot} (equation 7) and thus the inventories. Nevertheless, the advantage of equation 13 is that the majority of the terms can be directly quantified (*e.g.*, $F^{10\text{Be}}$, M_{tot} , r_{reac} , or $[{}^9\text{Be}]_{\text{reac}}$) allowing the primary mineral dissolution rate ($k_w X_{\text{prim}} M_{tot}$) to be evaluated. The partitioning term for the fraction of ${}^9\text{Be}$ incorporated into secondary minerals, $f_{\text{sec}}^{9\text{Be}}$, can be determined based on the mass depletion of ${}^9\text{Be}$ in the reactive and mineral pools according to elemental mass transfer coefficients:

$$f_{\text{sec}}^{9\text{Be}} = \frac{\left(\frac{[{}^9\text{Be}]_{\text{reac}}}{[{}^9\text{Be}]_{\text{parent}}} \frac{[X_I]_{\text{parent}}}{[X_I]_{\text{tot}}} \right)}{\left(1 - \frac{[{}^9\text{Be}]_{\text{min}}}{[{}^9\text{Be}]_{\text{parent}}} \frac{[X_I]_{\text{parent}}}{[X_I]_{\text{tot}}} \right)} \quad (14)$$

where $[X_I]_{\text{par}}$ and $[X_I]_{\text{tot}}$ are the concentrations of an immobile element in the parent material and the regolith, respectively, and the $[^9\text{Be}]_{\text{min}}$ primary mineral-bound ^9Be remaining after removal of all $^9\text{Be}_{\text{reac}}$ by a sequential extraction step (Wittmann et al. 2012).

The general features of **Fig. 11b**, where the $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ is compared to W , are consistent with equation 13. If $F^{10\text{Be}}$ is held constant, the $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ decreases as the weathering flux increases the amount of ^9Be accumulating in the weathering zone. According to our simulations (e.g., Fig. 9a and 9b), $I_{9\text{Be},\text{reac}}$ grows linearly over time and $I_{10\text{Be},\text{reac}}$ is nearly linear. This linear increase in both isotope inventories implies that the $dr_{\text{reac}}/dt \sim 0$ once the accumulation rates are constant. This isotopic steady state is attained very early (*ca.* 20 kyr) in the model scenarios. A version of equation 13 for isotopic steady state is:

$$r_{\text{reac},ss} = \frac{F^{10\text{Be}}}{\left(f_{\text{sec}}^{9\text{Be}} k_w X_{\text{prim}} M_{\text{tot}} [^9\text{Be}]_{\text{parent}} + L^{10} I_{9\text{Be},\text{reac}} + I_{9\text{Be},\text{reac}} \right)} \quad (15)$$

This would be the maximum $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ that could be achieved for set values of $k_d X_{\text{prim}} M_{\text{tot}} [^9\text{Be}]_{\text{parent}}$. We can also re-arrange to solve for W at isotopic steady state:

$$W = (1 - f_{\text{sec}}) k_w X_{\text{prim}} M_{\text{tot}} = \frac{1 - f_{\text{sec}}}{f_{\text{sec}}^{9\text{Be}} [^9\text{Be}]_{\text{parent}}} \left(\frac{F^{10\text{Be}}}{r_{\text{reac},ss}} - L^{10} I_{9\text{Be},\text{reac}} - I_{9\text{Be},\text{reac}} \right) \quad (16)$$

A key advantage of this particular formulations is that the application of $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ does not require knowledge of the erosion rate to determine the weathering rate.

4.2.2 $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ applied to steady-state weathering

Equations 13-16 represent a simple framework compared to the complexity represented in the model, which is still simple relative to actual weathering environments. To evaluate the ability of the equations to capture the processes occurring in the model, we consider the simplest case first assuming that $L^{10} = \lambda \approx 0$. This results in a simplified set of equations that require knowledge of X_{prim} and M_{tot} at a given time (t):

$$r_{\text{reac}} = \frac{F^{10}}{f_{\text{sec}}^{9\text{Be}} k_w X_{\text{prim}}(t) M_{\text{tot}}(t) [^9\text{Be}]_{\text{parent}}} \left(1 - \exp \left[\frac{-f_{\text{sec}}^{9\text{Be}} k_w X_{\text{prim}}(t) [^9\text{Be}]_{\text{parent}}}{[^9\text{Be}]_{\text{reac}}} t \right] \right) \quad (17)$$

Equation (17) also assumes that the initial $I_{10\text{Be}}$ is zero. The steady-state version, applicable when both isotope inventories increase at constant rates, is given by:

$$r_{\text{reac}} = \frac{F^{10\text{Be}}}{f_{\text{sec}}^{9\text{Be}} k_w X_{\text{prim}} M_{\text{tot}} [^9\text{Be}]_{\text{parent}}} \quad (18)$$

In **Fig. 11b** the results of the reactive transport model simulations are compared to calculated values of the $(\text{I}_{10\text{Be}}/\text{I}_{9\text{Be}})_{\text{reac}}$ assuming the $[^9\text{Be}]_{\text{parent}}$ value of 2.8×10^{-7} mol/g_{prim} and $f_{\text{sec}}^{9\text{Be}}$ of 1 using equation 17. Contours of $f_{\text{sec}}^{9\text{Be}}$ are provided for the lower retention cases (hollow symbols). Only at very low weathering rates of $<10 \text{ g km}^2 \text{ yr}^{-1}$ ($=10 \text{ t km}^2 \text{ yr}^{-1}$) does Be loss introduce a major uncertainty into estimates of W . At higher W , the $(\text{I}_{10\text{Be}}/\text{I}_{9\text{Be}})_{\text{reac}}$ faithfully records the weathering rate. In general, equation 18 provides a close match to the simulated $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$. In order to evaluate the utility of the steady-state approximation, the tabulated reactive transport results were used to compare equations 17 and 18 (**Fig. 11c**). Under the conditions represented by the model (*i.e.*, the term in the exponential is sufficiently large), the consequences of assuming steady state are minimal, meaning that equation 18 (no loss) is likely to be applicable to determining weathering rates in a variety of systems. The time to steady-state can also be observed by the change in slopes of the $(\text{I}_{10\text{Be}}/\text{I}_{9\text{Be}})_{\text{reac}}$ in **Fig. 9c** and is generally within 25 kyr.

The equations we present here are similar to those presented previously in equations 8 and 9 of von Blanckenburg et al. (2012) for steady-state weathering (*i.e.*, $E + W = RP = D$), implying a constant thickness of the weathering zone. In this approach, the $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ of river sediment (or water) was related to the ratio between F^{10} and the product of the total denudation rate ($D = E + W$), $[^9\text{Be}]_{\text{parent}}$, and the fractional fluxes ($f_{\text{reac}}^9 + f_{\text{diss}}^9$) of that portion of ^9Be that is released from primary minerals and mixes with ^{10}Be :

$$r_{\text{reac}} = \frac{F^{10}}{D[^9\text{Be}]_{\text{parent}}(f_{\text{reac}}^9 + f_{\text{diss}}^9)} = \frac{F^{10} J_{\text{riv}}^{9\text{Be}}}{D[^9\text{Be}]_{\text{parent}}(J_{\text{riv_reac}}^{9\text{Be}} + J_{\text{riv_diss}}^{9\text{Be}})} \quad (19)$$

where $J_{\text{riv}}^{9\text{Be}}$, $J_{\text{riv_reac}}^{9\text{Be}}$, and $J_{\text{riv_diss}}^{9\text{Be}}$ are the total, reactive and dissolved riverine fluxes of Be, respectively. This above equation can be simplified if we assume that that $J_{\text{riv}}^{9\text{Be}} = D[^9\text{Be}]_{\text{parent}}$:

$$r_{\text{reac}} = \frac{F^{10}}{(J_{\text{riv_reac}}^{9\text{Be}} + J_{\text{riv_diss}}^{9\text{Be}})} = \frac{F^{10}}{k_w X_{\text{prim}} M_{\text{tot}} [^9\text{Be}]_{\text{parent}}} \quad (20)$$

where the ratio of fluxes after the second equivalence is identical to equation (18) at full retention $f_{\text{sec}}^{9\text{Be}} = 1$. The weathering inventory model of equation (18) describes the $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$

ratio of inventories as a function of the gross primary mineral dissolution rate. In contrast, equation (19) describes the $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ ratio of eroded topsoil carried as river sediment and equilibrated with dissolved river Be, that tracks the denudation rate of soil or catchments von Blanckenburg et al., 2012). The agreement emerges from the steady state assumptions made in both approaches. In the weathering inventory model, equation 18 assumes that the $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ ratio of the isotope inventories that does not change with time (**Fig. 10b, 11c**) because both isotopes are entering the weathering zone at constant rates. The ratio thus tracks the rate of regolith production and the dissolution rate of primary minerals and is proportional to the (assumed constant) deepening rate of the weathering front. In equation (19), the flux of the two isotopes entering the weathering zone is assumed constant, such that the ^9Be inventory also tracks the deepening of the weathering front. The additional assumption of constant weathering zone thickness (von Blanckenburg et al, 2012) also implies that weathering front propagation rate equals the denudation rate. Hence, even though equation (18) describes a ratio of inventories whereas equation (19) relates the $^{10}\text{Be}/^9\text{Be}$ of river sediment (or river water) to the ratio in surface soil, the two sets of equations provide a consistent set of approaches for evaluating denudation rates derived from river sediment (or river water) and weathering rates in the regolith.

5. Conclusion

We suggest that the $^{10}\text{Be}/^9\text{Be}$ ratio provides a chronometer for determining both the ages of weathering zones and the long-term chemical weathering rates that sculpted them. The combination of reactive transport and differential mass balance approaches presented here suggests that when Be retention is high, the inventory of meteoric ^{10}Be in weathering zone profiles records the surface age. At ^9Be loss fractions less than approximately <15%, ^9Be loss provides an estimate of the maximum possible amount of ^{10}Be loss. This loss fraction can be used to correct for loss from the ^{10}Be inventory. The error on such corrections is lower for young surfaces due to more similar loss fractions between the two isotopes in less weathered material. The method thus holds substantial promise for dating of late Quaternary geomorphic features.

Our results also confirm that the mixing of ^9Be from weathering with ^{10}Be deposited at the Earth's surface provides a useful approach for constraining long-term weathering rates, even under cases of incomplete Be retention. The close agreement between the reactive transport

model and the analytical solutions (equations 18 and 19) suggest that weathering rates can be reliably calculated from the ratio of inventories of $^{10}\text{Be}_{\text{reac}}$ and $^9\text{Be}_{\text{reac}}$ ($I_{10\text{Be}}/I_{9\text{Be}})_{\text{reac}}$. Thus, an advantage of the coupled $^{10}\text{Be}/^9\text{Be}$ approach is that long-term weathering rates can be compared to other geochemical or isotopic methods using a relatively simple set of equations. Collectively, our results suggest that measurement of $^{10}\text{Be}/^9\text{Be}$ in weathered regolith provides a powerful tool for determining both surface ages and chemical weathering rates.

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Figure Captions

Figure 1. Conceptual model for reactive transport simulations of beryllium isotopes during weathering. (a) Abundance of primary and secondary minerals, (b) Input of meteoric ^{10}Be to the top of the weathering zone and concentrations of accumulated “reactive” ^{10}Be on secondary minerals in bulk regolith material, (c) Input of weathered ^9Be from meteoric deposition and

weathering of primary minerals, and concentrations of “reactive” ^9Be retained on secondary minerals and of the remaining ^9Be in primary minerals in bulk regolith material.

Figure 2: Comparison between measured partition coefficients, or K_d values, and the surface complexation description of Be sorption on secondary minerals (kaolinite here) using the fluid-to-solid ratio and total Be concentrations from the experiments of You et al. (1989); (solid symbols). These values are compared to other measured values from river sediment (Brown et al., 1992) and suspended sediment (Wittmann et al., 2015). Given the spread in the existing K_d data, site densities of between $1 \times 10^{-7} \text{ mol/m}^2$ and $1 \times 10^{-9} \text{ mol/m}^2$ bracket the data. We assume the site density of $5 \times 10^{-8} \text{ mol/m}^2$ (line 2) as the reference simulation corresponding to complete Be retention, and use the lower site densities to evaluate the loss fraction for both ^9Be and ^{10}Be . In the weathering simulations the K_d value is time- and depth-dependent due to changes in surface area.

Figure 3: Depth profiles for minerals (a-c, e-g) and porosity (d, h) as a function of time for a low (0.1 m/yr) infiltration flux (upper row, a-d) and higher (0.5 m/yr) infiltration flux (lower row, e-g). Note the shift in time intervals after 100 kyr.

Figure 4: Depth profiles of K_d values calculated from the surface complexation model with respect to time for bulk regolith at (a) high retention according to the reference simulation (site density = $5 \times 10^{-8} \text{ mol/m}^2$) and (b) for lower retention (site density = $5 \times 10^{-9} \text{ mol/m}^2$). The box plots at the bottom of each panel indicate the distribution of K_d values (median and upper and lower quartiles) from Figure 2 between pH 5 and 7 (mean is $2 \times 10^5 \text{ ml/g}$). Legend is as in Figure 3.

Figure 5: Depth profiles for $\text{SiO}_2(\text{aq})$, pH and $[^9\text{Be}]_{\text{diss}}$ and $[^{10}\text{Be}]_{\text{diss}}$ for a low flow rate of 0.1 m/yr (upper row, a-d) and high flow rate of 0.5 m/yr (lower row, e-g). Legend is as in Figure 3.

Figure 6: Depth profiles for sorbed Be species at low flow rate (0.1 m/yr) along the top row (a-c) and high flow rate (0.5 m/yr) for the bottom row (d-f). The $[^9\text{Be}]_{\text{reac}}$ and $[^{10}\text{Be}]_{\text{reac}}$ is calculated according to the total volume of minerals (*i.e.*, secondary plus primary minerals), (a, b, d, e). The $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ is the ratio of the sorbed concentrations (c, f) and the stippled line shows the corresponding detection limit ($^{10}\text{Be}/^9\text{Be} = 10^{-15}$) for accelerator mass spectrometry (AMS). The $[^{10}\text{Be}]_{\text{reac}}$ axis is truncated at the detection limit of 10^{-15} mol/m^3 (corresponding to ca. 10^4 atoms of ^{10}Be) for AMS.

Figure 7: Depth profiles of $[^{10}\text{Be}]_{\text{reac}}$ and corresponding inventory ($I_{^{10}\text{Be},\text{reac}}$) as a function of four flow rates over 200 kyr of model time at (a) high retention according to the reference simulation (site density = $5 \times 10^{-8} \text{ mol/m}^2$) and (b) for lower retention (site density = $5 \times 10^{-9} \text{ mol/m}^2$). In (a) the inventory is shown at an arbitrary depth with colors corresponding to the flow rate. In (b), the corresponding inventories are shown again at arbitrary depths with colors corresponding to the flow rates in (a). The numbers in parentheses indicate that corresponding age calculated from the time-integrated inventories. The grey line in both panels corresponds to the expected value of the inventory in the absence of ^{10}Be loss (*i.e.*, closed system). The inventory is calculated over the depth of the soil where porosity is <1% greater than the starting value as the depth of the weathering zone always exceeds the depth of the Be profiles in the model.

Figure 8: Depth profiles of $(^{10}\text{Be}/^9\text{Be})_{\text{reac}}$ and corresponding inventories $(I_{^{10}\text{Be}}/I_{^9\text{Be}})_{\text{reac}}$ as a function of flow rates over 200 kyr of model time at (a) high retention according to the reference simulation (site density = $5 \times 10^{-8} \text{ mol/m}^2$) and (b) for lower retention (site density = $5 \times 10^{-9} \text{ mol/m}^2$). The inventory is shown for arbitrary depths with colors corresponding to the flow rates. In (b), the closed symbols show the corresponding values for high Be retention for reference from (a) and open symbols show the effect of low retention on the $(I_{^{10}\text{Be}}/I_{^9\text{Be}})_{\text{reac}}$ due to preferential loss of ^9Be .

Figure 9. Time series for Be isotope inventories assuming different retentivities (*i.e.*, site densities) in the weathering zone for 0.1 m/yr infiltration. The site densities correspond to those of **Figure 2**. (a) $I_{^{10}\text{Be},\text{reac}}$ as a function of time compared to the analytical solution for the ^{10}Be inventory assuming a closed system with only ^{10}Be deposition and decay (equation 1), (b) $I_{^9\text{Be},\text{reac}}$ as a function of time, (c) The ratio of the two inventories, $(I_{^{10}\text{Be}}/I_{^9\text{Be}})_{\text{reac}}$, in the weathering zone over time (d) Comparison between the loss fraction for ^{10}Be and ^9Be showing the preferential loss of ^9Be over ^{10}Be . Time intervals used are from Fig. 3.

Figure 10: (a) Theoretical error on ^{10}Be surface ages due to loss of ^{10}Be (does not include analytical error); the maximum ^{10}Be loss is given in parentheses for the corresponding curves for the arid ($q = 0.01 \text{ m/yr}$) and humid case ($q = 0.5 \text{ m/yr}$). The values in parentheses indicate the site density as in Fig. 2 (*i.e.*, 2, 5×10^{-8} ; 3, 2.5×10^{-8} ; 4, $1 \times 10^{-8} \text{ mol/m}^2$). (b) Theoretical error on ^{10}Be surface age associated with correcting the $I_{^{10}\text{Be},\text{reac}}$ using the ^9Be loss fraction. The

maximum ^9Be loss is given in percentages near the corresponding curves for each site density for the same cases in panel (a).

Figure 11: (a) Relationship between net weathering rate (in $\text{g m}^{-2} \text{ yr}^{-1} = \text{t km}^{-2} \text{ yr}^{-1}$), as indicated by elemental depletion in the solid profiles or equivalently, the solute fluxes, and gross primary mineral dissolution ($k_d X_{\text{prim}} M_{\text{tot}}$). The slope of the line reflects the fraction of secondary minerals accumulating ($1 - f_{\text{sec}}$) according to equation 8 and suggests a constant value of 0.47. (b) The inventory $(I_{10\text{Be}}/I_{9\text{Be}})_{\text{reac}}$ as a function of the weathering flux for all four flow rates. The open symbols show the results for lower retention simulations. The heavy dashed line corresponds to equation (18) assuming $f_{\text{sec}}^{9\text{Be}} = 1$, implying full retention; light stippled lines correspond to less efficient retention as indicated. (c) comparison between the non-steady-state solution (equation 17) and the steady-state solution (equation 18).

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Table 1: Initial and boundary conditions used in simulations.**Initial system parameters**

Flow rates:	0 to 1	m/yr	
Temperature:	15	°C	
Flux ^{10}Be :	1.66×10^{-14}	mol/m ² /yr	a
	1.00×10^6	atoms/cm ² /yr	
Flux of ^9Be	1.00×10^8	mol/m ² /yr	b
Porosity	0.15	m ³ /m ³	

Initial (infiltrating) fluid composition:

<i>Species</i>	<i>Concentration (mol/L)</i>	
CO ₂ (g)	0.005	c
$^9\text{Be}^{2+}$	variable	d
$^{10}\text{Be}^{2+}$	variable	d
Na ⁺	1.00×10^{-5}	
Mg ²⁺	1.00×10^{-5}	
K ⁺	1.00×10^{-5}	
Ca ²⁺	9.00×10^{-5}	
Al ³⁺	1.65×10^{-10}	
SiO ₂ (aq)	1.00×10^{-7}	
Cl ⁻	1.20×10^{-4}	
NO ₃ ⁻	1.00×10^{-4}	
pH	5	

Initial solid compositions:

Species	^9Be conc.	Volume fraction	Surface area	Log rate constant
	(ppm)	(m ³ /m ³)	(m ² /g)	(mol/m ² /sec)
Albite(An ₂₀)	2.5	0.17	0.1	-14.0
K-feldspar	2.5	0.17	0.1	-14.6
Quartz	0	0.5	1	e
Halloysite	0	0.01	20	-18

a: mid-latitude average from Willenbring and von Blanckenburg, 2010.

b: determined from concentrations in average precipitation (Vesely et al., 2002).
corresponds to 1×10^{-10} mol/L at infiltration flux of 0.1 m/yr.

c: in bars

d: concentrations are adjusted at each infiltration flux to result in the same flux and $^{10}\text{Be}/^9\text{Be}$ ratio in each simulation.

e: assumed to be non-reactive (*i.e.*, dissolution rate is zero).

Table 2. Surface complexation reactions and parameters determined using CrunchFlow.

	Surface Reaction	Log K
1	$>(s)O^- + H^+ \leftrightarrow >(s)OH$	8.93 ^a
2	$>(s)OH + H^+ \leftrightarrow >(s)OH_2^+$	-7.29 ^a
3	$>(s)OBe^+ + H^+ \leftrightarrow >(s)H + Be^{2+}$	5.73 ^b

(a) Dzombak and Morel (1990) for an Fe-oxide surface site.
(b) Estimated by Dzombak and Morell (1990; Chap. 10, p. 299).

Table 3. Definition of model parameters.

Parameter	Definition	Values	Units
ϕ	Porosity		m^3/m^3
q	Flow rate of water through regolith	0.1 to 1	m/yr
M_i	Mass of material in the weathering zone (secondary (<i>sec</i>), primary (<i>prim</i>) or total (<i>tot</i>) ^a		g/m^2
$V_{m,j}$	Mineral volumes ^a		m^3/m^3
A_j	Bulk surface area		m^2/m^3
C_i	Aqueous concentration		mol/L
$S_{j,i}$	Sorbed concentrations		mol/m^3
E	Erosion rate		$\text{g}/\text{m}^2/\text{yr}$
RP_{prim}	Regolith production rate		$\text{g}/\text{m}^2/\text{yr}$
W	Net chemical weathering rate determined from solid profiles or solute fluxes, respectively		$\text{g}/\text{m}^2/\text{yr}$
k_w	Dissolution time constant for mineral dissolution		yr^{-1}
$k_w X_{prim} M_{tot}$	Gross primary mineral dissolution rate		$\text{g}/\text{m}^2/\text{yr}$
f_{sec}	Mass fraction of secondary minerals formed from primary mineral dissolution	0.47	-
$(1 - f_{sec})k_w X_{prim} M_{tot}$	Net chemical weathering rate (W) based on amount of primary mineral (X_{prim}) in weathering zone.		$\text{g}/\text{m}^2/\text{yr}$
$F^{10\text{Be}}$	Depositional flux of meteoric ^{10}Be	1.7×10^{-14}	$\text{mol}/\text{m}^2/\text{yr}$
$F^{9\text{Be}}$	Depositional flux of ^9Be	1.0×10^{-8}	$\text{mol}/\text{m}^2/\text{yr}$
$[^9\text{Be}]_{\text{par}}$	Concentration of ^9Be in parent material	2.8×10^{-7} (2.5 ppm)	$\text{mol}/\text{g}_{\text{parent}}$
$[^9\text{Be}]_{\text{min}}$	Concentration of primary ^9Be in weathering zone		$\text{mol}/\text{g}_{\text{tot}}$
$[^9\text{Be}]_{\text{diss}}$ or $[^{10}\text{Be}]_{\text{diss}}$	Concentration of dissolved ^9Be or ^{10}Be		mol/L
$[^9\text{Be}]_{\text{reac}}$ or $[^{10}\text{Be}]_{\text{reac}}$	Concentration of reactive ^9Be or ^{10}Be (<i>i.e.</i> , sorbed and co-precipitated)		$\text{mol}/\text{g}_{\text{tot}}$
$(^{10}\text{Be}/^9\text{Be})_{\text{reac/diss}}$	Isotope ratio of ^9Be to ^{10}Be in reactive or dissolved pool (abbreviated r_{reac})		-

$I_{10\text{Be, reac}}$	Inventory of ^{10}Be in reactive pool	mol/m^2
$I_{9\text{Be, reac}}$	Inventory of ^9Be in reactive pool	mol/m^2
$(I_{10\text{Be}}/I_{9\text{Be}})_{\text{reac}}$	Ratio of the inventories of ^9Be and ^{10}Be	-
K_d	Solid/fluid partition coefficient	ml/g
$f_{\text{sec}}^{9\text{Be}}$	Fraction of weathered ^9Be retained in secondary minerals	-
L^{10}	Loss rate of ^{10}Be	yr^{-1}
λ	Decay constant for ^{10}Be	$4.9975 \times 10^{-7} \text{ yr}^{-1}$

^a index j refers to the solid phase and i refers to the chemical component

^{10}Be (meteoric) and ^9Be during weathering

Relationship between sorbed $^{10}\text{Be}/^9\text{Be}$ and weathering rate