INTRODUCTION

Theories that link atmospheric and geological processes (Berner, 1994; Raymo and Ruddiman, 1992; Urey, 1952) have stimulated numerous studies that have explored the interactions among climate, erosion, and chemical weathering (e.g., Oliva et al., 2003; Riebe et al., 2004; West et al., 2005; White and Blum, 1995). At the heart of these theories is the common understanding that weathering of silicate minerals consumes atmospheric CO$_2$ (Urey, 1952). The identification of the dominant control on chemical weathering, however, has been a source of debate. Some advocate that climate, in particular, temperature, is the critical factor, such that an increase in atmospheric CO$_2$ would raise global temperatures, thus accelerating weathering rates (e.g., Kump et al., 2000; White and Blum, 1995). The increase in weathering rates, in turn, would draw down atmospheric CO$_2$, thus lowering temperatures through a negative feedback process (Berner et al., 1983; Walker et al., 1981). In contrast, others propose that orogenetic activity enhances erosion rates, thus exposing fresh minerals to chemical weathering processes (Raymo and Ruddiman, 1992; Raymo et al., 1988). The increased supply of fresh minerals drives higher chemical weathering rates, thus drawing down atmospheric CO$_2$ and lowering temperatures.

Although the debate is not yet settled, there is growing evidence that erosion may play an important role in controlling chemical weathering rates (Gaillardet et al., 1999; Millot et al., 2002; Riebe et al., 2001, 2004; West et al., 2005). However, it is the precise nature of the dependency of weathering on erosion that determines its sensitivity to changes in erosion rate. The relationship between erosion rate ($E$) and weathering rate ($W$) can be simply expressed with: $W \propto E^\lambda$, where $\lambda$ is a dimensionless constant. If $\lambda \approx 1$, increases in erosion will be matched by similar increases in weathering, but if $\lambda < 1$, large increases in erosion will not lead to large increases in weathering, even where erosion is the dominant control on weathering. From empirical studies, values of $\lambda$ include ~1 (Riebe et al., 2001, 2004; West et al., 2005), 0.66 (Millot et al., 2002), and 0.42 (West et al., 2005).

To explain the range of $\lambda$ values, Riebe et al. (2004) and West et al. (2005) proposed that in landscapes with a limited supply of fresh minerals (i.e., supply-limited), the relationship between erosion and chemical weathering should be linear. However, in landscapes with sufficiently high erosion rates, weathering will be limited by the kinetics of chemical reactions, and $\lambda$ will be less than unity (Riebe et al., 2004; West et al., 2005). To formalize these distinctions, West et al. (2005) introduced a set of equations relating weathering rates to erosion rates for the two end-member weathering regimes. Inspired by West et al.’s (2005) approach, we present a theoretical model that encompasses the continuum of weathering conditions, from supply-limited to kinetically limited. This model allows us to evaluate the sensitivity of changes in chemical weathering rates to changes in erosion rates.

MODEL

Formulation

The model is based on a mass-balance approach in which material enters the weathering zone through the downward propagation of the weathering front and leaves the weathering zone through physical erosion at the land surface and through the transport of dissolved phases by chemical weathering (Fig. 1). By geochemical convention, this material is defined as regolith and includes both a zone of only chemically altered material and a zone of physically disturbed material (e.g., Yoo and Mudd, 2008a). Assuming steady-state regolith thickness, the weathering rate per land surface area ($W/\text{M L}^{-2} \text{T}^{-1}$) can be expressed as

$$W = \frac{m_c - m_b}{\Delta t},$$

where $m_c$ (M L$^{-2}$) is the mass per land surface area of weatherable minerals entering the column through the advance of the weathering front, $m_b$ (M L$^{-2}$) is the mass of weatherable minerals leaving through erosion, and $t$ is time. Refractory minerals do not contribute to the weathering flux and thus are not included in Equation 1. Equation 1 is an expression of mass balance whereby the spatially integrated weathering rate of the weathering column (or, alternatively, the mass lost in solution) is the total out to the adjacent landscape. This model extends the mass balance to the adjacent landscapes.

Figure 1. In a steadily eroding landscape, material entering control volume ($m_c$) is balanced by material leaving in solution form ($W$) and by physical erosion ($m_b$).
difference between the rate of unweathered material entering the weathering column via bedrock erosion and the rate of weathered material being removed by physical erosion. The input term, \( m_c \), is defined as:

\[
    m_c = E \chi_m \Delta t,
\]

where \( E \) is the weathering advance rate (\( M L^{-2} T^{-1} \)), and \( \chi_m \) is the mass fraction of chemically mobile material (i.e., \( 1 - \chi_m \) is the fraction of chemically inert material). In a steadily eroding and weathering landscape, the erosion rate will be equal to the downward propagation of the weathering front.

Several studies have found that the weathering rates of primary minerals are inversely proportional to the amount of time the minerals spend in the weathering zone (e.g., Hodson and Langan, 1999; White and Brantley, 2003). Compiling data from a large number of studies, White and Brantley (2003) found that the weathering rate can be expressed as a power-law function of the time minerals have spent in the weathering zone. Yoo and Mudd (2008b) demonstrated that, in order to apply weathering rates obtained from noneroding surfaces (e.g., many of the studies compiled by White and Brantley, 2003) to an erosional setting, one must account for the chemical evolution of individual minerals as opposed to a bulk “age” of the weathering zone. We apply this approach such that the loss rate of mass per area (\( m \)) by chemical weathering is expressed as:

\[
    \frac{dm}{dt} = -mK\rho^\sigma,
\]

where \( K (T^{-1}) \) and \( \sigma \) (unitless) are empirically derived constants (see Appendix DR1 in the GSA Data Repository). The weathering rate constant \( K \) used throughout this derivation is not the same rate constant presented in White and Brantley (2003), although they are functionally related (Yoo and Mudd, 2008b). We note that the extent of weathering of an individual mineral that moves up from the weathering front, through the weathering zone, and eventually to the surface, is time-dependent. If the erosion rate is steady, however, the amount of time particles spend in the weathering zone is constant; thus, by integrating Equation 3, we can quantify the mass remaining in minerals emerging from a steadily eroding landscape:

\[
    m = m_c e^{-K\rho^\sigma \phi},
\]

where \( m_c \) is the original mass of the unweathered material, and \( T \) is the residence time of the material as it travels through the regolith. Following from Equations 2 and 4, the mass of weathered material leaving the regolith through physical erosion is expressed as

\[
    m_e = E \chi_m e^{-K\rho^\sigma \phi},
\]

Inserting Equations 2 and 5 into Equation 1, and simplifying, yields

\[
    W = E \chi_m \left( 1 - e^{-K\rho^\sigma \phi} \right).
\]

Assuming that physical erosion removes material from the top of the regolith surface, the residence time of the unit mass is calculated as

\[
    T = \frac{ph}{E},
\]

where \( p \) is the density (\( M L^{-3} \)), \( E \) is the erosion rate (\( M L^{-2} T^{-1} \)). Regolith thickness, itself, is strongly dependent on erosion; however, we are not aware of a comprehensive data set from which to derive a functional relationship between the two. The rate at which material is entrained into the physically disturbed portion of the regolith (often called soil production) has been found to decline exponentially with increasing depth of this disturbed layer (Heimsath et al., 1997), and the extent of weathering in the regolith below the physically disturbed layer has been found to covary with the rate of soil production (Burke et al., 2007). In addition, explicit modeling of geochemical processes in an eroding column suggests a nonlinear inverse relationship between regolith thickness and physical erosion rates (Lebedeva et al., 2007). Thus, we adopt a similar form such that

\[
    E = k_e e^{-\phi},
\]

where \( k_e \) and \( \phi \) are empirically derived constants. Solving Equation 8 for regolith depth yields

\[
    h = \frac{\ln(E/k_e)}{-\phi}.
\]

To determine the chemical weathering rate for a specified erosion rate, the regolith depth is calculated as a function of erosion rate with Equation 9. The regolith depth and the erosion rate are then used to calculate the residence time with Equation 7. Finally, the residence time and the erosion rate are used in Equation 6 to determine the weathering rate.

**Assumptions**

Equations 6, 7, and 9 define a quantitative model that formalizes the dependency of chemical weathering rates on erosion rates. As with all models, several assumptions were made in its derivation. First, we assume that erosion is steady, uniform, and occurs by processes that remove the top layer of regolith (i.e., the oldest, most weathered material). Although this would appear to limit the usefulness of this model in rapidly eroding landscapes where landsliding is dominant, results from Gabet (2007) suggest that this simplification of the erosional processes may yield results similar to a more elaborate treatment. Gabet (2007) derived a model that coupled chemical weathering processes with a stochastic representation of landslide magnitude and frequency and found the same relationship between weathering and erosion as West et al. (2005) did for the case of steady and spatially uniform erosion. On the basis of these results, the assumption of steady, uniform erosion made here may adequately represent a more complex reality.

Second, we assume that mixing within the entire regolith profile is insignificant. Mixing by processes such as bioturbation (Gabet et al., 2003) homogenizes the upper zone of the regolith such that particles eroding from the regolith surface have a distribution of ages, where young particles erode coevally with old particles (Almond et al., 2007; Mudd and Furbish, 2006; Yoo and Mudd, 2008b), whereas particles eroding from the top of an unmixed regolith have an age of \( T \) (i.e., the residence time). Thus, the mean age of fully mixed regolith is slightly younger than unmixed regolith and this will affect the estimate for the effective particle size (see following). In addition, the variance of particle age in a fully mixed regolith is equal to the square of the residence time, so discrepancies in predicted weathering rates in mixed versus unmixed regolith are minimized at high erosion rates (Mudd and Furbish, 2006).
which is difficult to constrain. We must, therefore, use particle size as a fitting parameter; its final value will be evaluated. Values for the constants defining the relationship between erosion rate and regolith depth ($k_h$ and $\phi$) determined in prior studies (e.g., Heimsath et al., 1997) are not appropriate in this context because those data do not span the range of erosion rates, and prior studies focused on physically disturbed material and not the entire regolith column. If Equation 9 is approximately appropriate, however, reasonable estimates based on known regolith depths at the limits of high and low erosion rates can be made for these parameters. At erosion rates on the order of $10^{-4}$ t km$^{-2}$ yr$^{-1}$, regolith depths may be of order 5 m (Bierman et al., 1995; White et al., 2002); note that we exclude non-eroding, lateritic landscapes. At erosion rates of $10^2$ t km$^{-2}$ yr$^{-1}$, the bedrock surface is being lowered at a rate of ~4 mm/yr; because this is faster than estimated rates of regolith production (e.g., Heimsath et al., 1997), average regolith depths must approach zero. To represent this very shallow weathering zone, we assume a depth of $10^{-2}$ m. For dimensional consistency, regolith depths were expressed in kilometers in Equation 9 to produce a value of $10^{5}$ t km$^{-2}$ yr$^{-1}$ for $k_h$ and $2300$ km$^{-1}$ for $\phi$. These estimated values are relatively insensitive to the minimum regolith depth: minimum depths of $10^{-1}$ m and $10^{-3}$ m produced nearly identical values of $k_h$ and $\phi$.

RESULTS AND DISCUSSION

As explained above, the effective diameter of the weathered particles ($D$; Appendix DR1) is the only parameter value that cannot be determined a priori. Weathering rates as determined by Equation 6, therefore, are calibrated to the data set compiled by West et al. (2005) for granitic landscapes. Because the data from West et al. (2005) include weathering rates from a range of rainfall and temperature regimes, their data were normalized by climatic conditions to isolate erosion as the sole control on weathering rate (see Appendix DR2). Guided by the minimum root mean square error, we find that, with a value of 28 $\mu$m for the effective particle size, Equation 6 predicts weathering rates remarkably well (Fig. 2); this particle size implies that $K = 0.0032$ yr$^{-1}$. The effective particle size is within the range of sizes compiled by White and Brantley (2003), particularly for conditions of rapid weathering and short residence times. To assess the sensitivity of the relationship between denudation and weathering on effective particle diameter, chemical weathering rates were predicted with an order of magnitude increase and decrease in $D$; these new values form a tight envelope around the field data (Fig. 2). In a fully mixed regolith, the average particle age would be younger and would therefore lead to a lower estimate for the particle size.

Under supply-limited conditions ($10^0$–$10^4$ t km$^{-2}$ yr$^{-1}$), the relationship between weathering and erosion is approximately linear. This can be appreciated analytically with Equation 6, whereby in the limit of low erosion rates ($E \to 0$), the residence time approaches infinity and $W = E \lambda$. This is a condition of complete weathering where the weathering rate is limited by the supply rate of weatherable material (Riebe et al., 2004; West et al., 2005).

As denudation increases, weathering enters the kinetically limited regime, and the relationship between erosion and weathering rate becomes less than linear. The deviation from the linear relationship is due to the tension between the positive effects of fresher (and thus more reactive) minerals and the negative effects of a thinner regolith. Because fresher minerals weather faster, the weathering rate per unit volume increases with denudation (Fig. 3). However, rapidly eroding landscapes have thinner regolith; thus, despite the increase in weathering rate per unit volume, the weathering rate per unit land surface area decreases. In the range of denudation rates of $10^{-3}$–$10^{-1}$ t km$^{-2}$ yr$^{-1}$, chemical weathering rates reach a plateau, and at the highest erosion rates (> $10^4$ t km$^{-2}$ yr$^{-1}$), weathering rates decline. Finally, Equation 6 predicts that solute fluxes from watersheds would be at a maximum where regolith thickness is ~0.50 m (Fig. 3, inset). The conflicting values of $\lambda$ found in the empirical studies mentioned earlier can now be reconciled. The form of the relationship between weathering and erosion (Fig. 2) demonstrates that $\lambda$ varies from 1 to 0 (even becoming negative at the extreme end of erosion rates), depending on where the sampled landscapes lie along the continuum between supply-limited and kinetically limited. In the studies where the erosion rates were dominantly in the range $10^0$–$10^3$ t km$^{-2}$ yr$^{-1}$, $\lambda$ is approximately unity (Riebe et al., 2001, 2004; West et al., 2005), and the weathering regime is supply-limited. In Millot et al.’s (2002) study, the rates range from $10^0$ to $5 \times 10^2$ t km$^{-2}$ yr$^{-1}$, thus extending the relationship into the margins of the kinetically limited regime and yielding a lower value for $\lambda$ ($0.66$). Finally, West et al.’s (2005) value of 0.42 for $\lambda$ comes from erosion rate data that span $10^{-10}$ t km$^{-2}$ yr$^{-1}$, thus sampling from a range of landscapes that are dominantly kinetically limited.
Our model (Fig. 2) and the field data (West et al., 2005) suggest that $\lambda$ is less than linear where denudation rates exceed $10^2$ km$^{-2}$ yr$^{-1}$, and it becomes vanishingly small (and eventually negative) as erosion rates increase; thus, the role of denudation and in situ chemical weathering in affecting the global climate becomes less and less important as erosion rates intensify. Denudation rates in the region of the High Himalayas, the location identified by Raymo and Ruddiman (1992) in their hypothesis relating erosion and weathering, would have driven the greatest increases in chemical weathering only during the earliest stages of orogeny. Presently, the High Himalayas have denudation rates in the range of $10^2$ to $10^4$ km$^{-2}$ yr$^{-1}$ (e.g., Blythe et al., 2007; Burbank et al., 2003), suggesting that in situ weathering rates there have peaked. The present role of erosion, however, in delivering fine-grained unweathered materials to the lowlands where they may undergo extensive weathering may be more important (Gaillardet et al., 1999). As shown in Equations 7 and 9, regolith thickness, residence time, and erosion rate are inextricably linked. At the highest erosion rates, the volumetric weathering rate is high, but, due to the thin regolith, the residence time is vanishingly small, and minerals leave the weathering zone virtually unaltered by weathering reactions. Indeed, high weathering rates have been measured on the Gangetic plain, where minerals deposited on floodplains have greater residence times (Derry and France-Lanord, 1997; Galy and France-Lanord, 1999).

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