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The Discrepancy between Mineral Residence Time and Soil Age: Implications for the Interpretation of Chemical Weathering Rates

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Derivation of Eq. 3

The mass loss rate of mineral species *j* per unit ground surface area via chemical weathering (w_j, with the unit of $ML^{-2}T^{-1}$) is described as:

$$w_j = -\frac{\partial m_j}{\partial \tau_j} = R_j A_{s,j} \omega_j . \qquad \text{(A)}$$

where m_j is the mass of mineral *j* per a given ground surface area [ML^{-2}], τ is the time that the minerals are exposed to chemical weathering reactions, R is the chemical weathering rate in moles reacted per mineral surface area per time [mol L^{-2} T^{-1}], A_s is the mineral surface area per ground surface area [unitless], and w_m is molar weight of the mineral j [M $mole^{-1}$], The surface area of the minerals per ground surface area [A_s, unitless] is related to the mineral grain roughness by [White and Brantley, 2003]:

$$A_{s,j} = \frac{6\lambda_j}{D_j \rho_{\mu,j}} m_j, \tag{B}$$

where D is the grain diameter [L], λ is the mineral surface roughness [unitless], and ρ_m is the density of the minerals [ML^{-3}].

Both the mass and the surface area of the minerals may evolve due to chemical weathering. In the model presented here we adopt the *White and Brantley* [2003] weathering model, which describes the chemical weathering rate per mineral surface area and the mineral grain roughness as a function of the mineral's exposure time to chemical weathering as

$$R_j = a_j \tau_j^{\alpha_j}, \tag{Ca}$$

and

$$\lambda_j = b_j \tau_j^{\beta_j}, \tag{Cb}$$

where *a, b, α,* and *β* are mineral species specific empirical coefficients.

Thus the mass loss rate of mineral species *j* per ground surface area via chemical weathering (w_j in Eq. A) can be rewritten as in the Eq.3 in the text:

$$w_j = -\frac{\partial m_j}{\partial \tau} = \underbrace{\frac{6 a_j b_j \omega_j}{D_j \rho_{\mu,j}} \tau_j^{\alpha_j + \beta_j}}_{\substack{time-dependent \\ rate\ coefficient\,[yr^{-1}]}} \times \underbrace{m_j}_{current\ mass} . \quad \text{(D)}$$

This equation describes a first order decay with time-dependent decay rate; the rate W_j is the product of the time-dependent rate coefficient and the current mass of *j*. If the initial mass per area of the mineral *j* is $m_{j,0}$, the m_j in Eq. D can be solved as a function of weathering exposure time (τ) and the initial mass ($m_{j,0}$):

$$m_j = m_{j,0} \exp[-\frac{6 a_j b_j \omega_j}{D_j \rho_{\mu,j} (1 + \alpha_j + \beta_j)} \tau_j^{1+\alpha_j+\beta_j}] . \quad \text{(E)}$$

Derivation of Eq. 4:

Over the course of soil formation, primary mineral grains of different species are continuously incorporated into the soil. The amount of mineral, *j*, per unit ground surface area that enters the soil from the underlying fresh parent material via soil production between the time period of *t* and *t+dt* [$m_{j,t}$ with the unit of ML^{-2}], where the t is the soil age, is:

$$m_{j,t} = (\rho_r C_{j,r} P_t)dt \text{ ,} \tag{F}$$

where ρ_r is the bulk density of the parent material [ML^{-3}], C_{jr} is the mass concentration of the primary mineral j in the parent material [MM^{-1}], and P_t is the soil production rate at time t [LT^{-1}].

Now we consider a soil of age T. As described in the time-dependent dissolution rate law described in Eq. D, the mass loss rate via chemical weathering from the mass fraction, $m_{j,t}$ in Eq. F, is the product of (1) the time-dependent reaction coefficient for the weathering exposure time of T-t and (2) the present mass of j that entered the soil between t and t+Δt and has survived the chemical weathering loss until time T (i.e., $m_{jt\rightarrow T}$).

$$w_{j,t,T} = \frac{6a_j b_j \omega_j}{D_j \rho_{\mu,j}} (T-t)^{\alpha_j + \beta_j} \times m_{j,t\rightarrow T} \text{ .} \tag{G}$$

As was done for Eq. E, $m_{j,t\rightarrow T}$ can be solved as a function of initial mass ($m_{j,t}$ in Eq. F) and the residence time, T-t.

$$m_{j,t\rightarrow T} = (\rho_r C_{j,r} P_t)dt \ \exp[-\frac{6a_j b_j w_j}{D_j \rho_{\mu,j}(1+\alpha_j+\beta_j)} (T-t)^{1+\alpha_j+\beta_j}] \tag{H}$$

The soil of age T is comprised of primary minerals that have been incorporated into the soil during the entire duration of soil formation. The mass loss from the mineral species j in the entire soil profile ($w_{j,T}$ with the unit of $ML^{-2}T^{-1}$) can be thus obtained by integrating $w_{j,t,T}$ (Eq. G) over the time period from t=0 to t=T.

$$w_{j,T} = \int_{t=0}^{t=T} w_{j,t,T} \; dt , \qquad \text{(Ia)}$$

$$= \frac{6a_j b_j \omega_j}{D_j \rho_{\mu,j}} \int_{t=0}^{t=T} \left[(T-t)^{\alpha_j+\beta_j} (\rho_r C_{j,r} P_t) \exp[-\frac{6a_j b_j \omega_j}{D_j \rho_{\mu,j}(1+\alpha_j+\beta_j)} (T-t)^{1+\alpha_j+\beta_j}] \right] dt , \qquad \text{(Ib)}$$

$$= \frac{6a_j b_j \omega_j}{D_j \rho_{\mu,j}} \int_{t=0}^{t=T} \left[(T-t)^{\alpha_j+\beta_j} m_{j,t\to T} \right] dt , \qquad \text{(Ic)}$$

Eq. (Ic) follows from Eq. (Ib) by inserting Eq. (H) and is featured as Eq.4 in the text.

Derivation of Eq. 5

If we incorrectly equate the mineral residence time (τ=T-t) and soil age (T), as has been done in the past, the chemical weathering rate of mineral species j from a soil of age T is described in Eq.5 in the text:

$$w_{j,T} = \underbrace{\frac{6a_j b_j \omega_j}{D_j \rho_{\mu,j}} T^{\alpha_j + \beta_j}}_{\substack{time-dependent \\ rate\ coefficient\,[yr^{-1}]}} \times \underbrace{(\rho_s C_{j,s} h_s)}_{current\ mass}. \quad \text{(J)}$$

where the current mass of mineral j is calculated as the product of soil bulk density, mass concentration of j, and soil thickness at time T.

Simulation Procedure

In the simulations, mineral grains are introduced into the soil during each timestep. The mass per unit ground surface area of the mineral grains introduced during a timestep of time dt is

$$m_{j,0} = P\chi_j \rho_{\mu,j}(1-\phi)dt\,, \quad \text{(K)}$$

where P is the soil production rate, determined by the soil thickness (see text), χ_j is the volume fraction of the mineral in the parent material (mass fraction and volume fraction are assumed to be the same in the simulations presented here because quartz, plagioclase, and K-feldspar have similar mineral densities), ρ_μ is the mineral density of species j, and ϕ is the porosity, assumed to be 0.4 for all simulations. $\rho_\mu \chi_j(1-\phi)$ is identical to $\rho_r C_{jr}$ in Eq. (F). It should be noted that the simulated particles represent a cohort of individual mineral grains that are produced in the time period Δt. Each mineral grain is then tagged with an age; the clock starts once the mineral grain is introduced into the soil layer.

As time passes, the soil-profile-integrated mass and weathering rate of the mineral j is updated according to Eq. (I). Thus the mass in the total soil profile and also of individual mineral grains of a range of residence times is tracked. The soil thickness is determined by summing the contribution of all the mineral grains that reflect mineral species groups and number of mineral particles within each group:

$$h = \sum_{j}^{\#species} \sum_{n}^{\#particles} \frac{m_{j,n}}{\chi_{j,i} \rho_{\mu,j,n} (1-\phi)}. \quad \text{(L)}$$

This soil thickness is then used to determine P in the following time-step with Eqs. 2a or 2b.

A Note on Residence Time

In the simulations, we track the time lengths that individual mineral grains spend in the soil. The residence time is calculated based on the average of those time lengths. The mineral grains, however, may have different masses (due to, for example, differential weathering of varying soil production) such that the residence time is calculated by weighting the time lengths of the mineral grains by their masses.

Reference

White, A., and S.L. Brantley (2003), The effect of time on the weathering of silicate Minerals: why do weathering rates differ in the laboratory and field?, *Chemical Geology*, *202*, 479-506.