Strong tectonic and weak climatic control of long-term chemical weathering rates

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ABSTRACT

The relationships among climate, physical erosion, and chemical weathering have remained uncertain, because long-term chemical weathering rates have been difficult to measure. Here we show that long-term chemical weathering rates can be measured by combining physical erosion rates, inferred from cosmogenic nuclides, with dissolution losses, inferred from the rock-to-soil enrichment of insoluble elements. We used this method to measure chemical weathering rates across 22 mountainous granitic catchments that span a wide range of erosion rates and climates. Chemical weathering rates correlate strongly with physical erosion rates but only weakly with climate, implying that, by regulating erosion rates, tectonic uplift may significantly accelerate chemical weathering rates in granitic landscapes.

Keywords: geochemical mass balance, cosmogenic nuclides, chemical weathering rates, climate.

INTRODUCTION AND METHODOLOGY

Physical erosion and chemical weathering are interrelated processes (Stallard and Edmond, 1983) that together regulate soil depth and development, deliver sediment and solutes to aquatic habitats, and shape mountainous landscapes. Measuring how rates of physical erosion and chemical weathering interrelate is therefore important for quantitative pedology, land-use management, and landscape evolution modeling. Silicate weathering modulates ocean alkalinity and is thus a long-term sink for atmospheric CO_2 (e.g., Berner et al., 1983), which regulates climate via the greenhouse effect. Quantifying how climate, weathering, and erosion are related is therefore essential for understanding how geomorphology and tectonics affect Earth's long-term climatic evolution (Raymo et al., 1988; Molnar and England, 1990). To the extent that chemical weathering rates increase with temperature, weathering feedbacks should, over millions of years, buffer Earth's climate against large temperature shifts (e.g., Berner et al., 1983). To the extent that chemical weathering rates are strongly coupled with physical erosion rates (Stallard and Edmond, 1983), and thus with mountain uplift rates, periods of increased mountain uplift would be marked by global cooling, due to increased atmospheric CO_2 consumption by weathering (Raymo et al., 1988).

Previous Measurements of Chemical Weathering Rates

Relationships among chemical weathering, physical erosion, and climate have remained poorly quantified because long-term chemical weathering rates have been difficult to measure. Chemical weathering rates have typically been measured from year- to decade-long records of solute fluxes, but these short-term measurements do not necessarily apply on the long time scales over which soils, landscapes, and climates evolve. In the rare instances in which soil age can be determined and soil erosion can be assumed to be negligible, long-term chemical weathering rates can be measured using soil mass-balance techniques (April et al., 1986). According to the mass-balance approach, as the relatively soluble minerals in soils dissolve away, the more immobile elements should become increasingly enriched relative to their concentrations in unweathered parent material. Measurements of immobile element enrichment therefore reveal the degree of soil weathering, and can be used to quantify the total dissolution loss from a soil. The average weathering rate can then be estimated by dividing the dissolution loss by the soil age. However, because non-eroding soils of known age are rare, this mass-balance approach cannot be applied in many environments.

New Mass-Balance Approach for Measuring Weathering Rates

Here we show how the soil mass-balance approach can be extended to measure long-term weathering rates in eroding landscapes. If soil formation from rock is counterbalanced by soil loss from physical erosion and chemical weathering, then soil depth will be constant (e.g., Heimsath et al., 1997) and

$$P_{\text{soil}} = D = E + W, \tag{1}$$

where P_{soil} is the soil production rate, D is the total denudation rate, E is the physical erosion rate, and W is the chemical weathering rate. For insoluble elements like zirconium, removal by weathering should be zero, so inputs from soil production will be balanced solely by outputs from physical erosion:

$$[\text{Zr}]_{\text{rock}} \times D = [\text{Zr}]_{\text{soil}} \times E, \tag{2}$$

where $[\text{Zr}]_{\text{rock}}$ and $[\text{Zr}]_{\text{soil}}$ are representative zirconium concentrations of the rock and soil. By substitution, we can rewrite equation 1 as:

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TABLE 1. CHEMICAL DEPLETION FRACTIONS AND WEATHERING RATES*

Basin	Denud-ation rate† (t·km^{-2}·yr^{-1})	[Zr]$_{saprolite}$ (ppm)§	[Zr]$_{outcrop}$ (ppm)§	[Zr]$_{rock}$ (ppm)§	[Zr]$_{soil}$ (ppm)§	Chemical depletion fraction (%)	Weathering rate** W_{Si} (t·km^{-2}·yr^{-1})	W
Fort Sage (granodiorite; 25 ± 3 cm/yr; 12.2 ± 0.6 °C; average depletion = 15% ± 3%)								
A1	77 ± 7	N.D.	112 ± 3 (2)	112 ± 3	118 ± 5 (5)	6 ± 5	0 ± 1	4 ± 4
A2(s)	56 ± 15	120 ± 4 (5)	112 ± 3 (2)	118 ± 3	139 ± 5 (19)	15 ± 4	2 ± 1	9 ± 3
A3(s)	152 ± 27	N.D.	113 ± 4 (4)	113 ± 4	139 ± 7 (16)	18 ± 5	8 ± 3	28 ± 9
A4(s)	680 ± 211	118 ± 4 (4)	122 ± 3 (2)	119 ± 3	142 ± 3 (22)	16 ± 3	36 ± 13	110 ± 38
Fall River (tonalite; 145 ± 5 cm/yr; 11.9 ± 0.6 °C; 19% ± 2%)								
FR-2	393 ± 82	94 ± 9 (12)	85 ± 6 (3)	92 ± 7	109 ± 6 (13)	15 ± 8	19 ± 12	60 ± 34
FR-5	293 ± 36	121 ± 5 (7)	114 ± 3 (4)	119 ± 3	150 ± 5 (25)	21 ± 3	21 ± 4	61 ± 13
FR-6	89 ± 25	N.D.	83 ± 5 (3)	83 ± 5	102 ± 3 (9)	19 ± 5	6 ± 2	17 ± 7
FR-8	38 ± 5	N.D.	87 ± 1 (3)	87 ± 1	106 ± 3 (9)	18 ± 3	2 ± 1	7 ± 1
Adams Peak (granodiorite; 58 ± 7 cm/yr; 4.2 ± 0.5 °C; average depletion = 14% ± 2%)								
AP-3	124 ± 12	111 ± 4 (5)	112 ± 1 (3)	111 ± 2	135 ± 3 (20)	17 ± 2	6 ± 1	21 ± 4
AP-4	83 ± 8	N.D.	108 ± 1 (3)	108 ± 1	115 ± 4 (9)	6 ± 4	1 ± 1	5 ± 3
AP-5	148 ± 13	N.D.	112 ± 3 (4)	112 ± 3	129 ± 7 (9)	12 ± 6	5 ± 3	18 ± 8
AP-11	90 ± 12	N.D.	109 ± 6 (2)	109 ± 6	129 ± 5 (4)	15 ± 6	4 ± 2	14 ± 5
AP-13	117 ± 12	N.D.	112 ± 5 (2)	112 ± 5	131 ± 3 (4)	14 ± 4	5 ± 2	17 ± 5
Antelope Lake (granodiorite; 83 ± 6 cm/yr; 7.8 ± 0.4 °C; average depletion = 18% ± 5%)								
AL-4	66 ± 8	N.D.	166 ± 15 (4)	166 ± 15	227 ± 8 (9)	27 ± 7	5 ± 2	18 ± 5
AL-5	78 ± 12	N.D.	165 ± 48 (2)	165 ± 48	211 ± 10 (7)	22 ± 23	5 ± 5	17 ± 18
AL-9	98 ± 13	N.D.	211 ± 25 (3)	211 ± 25	271 ± 18 (8)	22 ± 11	6 ± 3	22 ± 11
AL-10	81 ± 8	182 ± 11 (8)	192 ± 7 (2)	183 ± 9	212 ± 6 (26)	14 ± 5	3 ± 1	11 ± 4
Sunday Peak (granite; 105 ± 5 cm/yr; 9.4 ± 0.4 °C; average depletion = 10% ± 4%)								
SP-1	108 ± 12	228 ± 16 (8)	214 ± 7 (3)	225 ± 12	252 ± 7 (23)	11 ± 6	4 ± 2	12 ± 6
SP-3	82 ± 10	N.D.	238 ± 12 (3)	238 ± 12	245 ± 16 (9)	3 ± 8	1 ± 2	3 ± 7
SP-8	82 ± 8	N.D.	244 ± 13 (4)	244 ± 13	278 ± 7 (8)	12 ± 5	3 ± 2	10 ± 4
Nichols Peak (granodiorite; 22 ± 3 cm/yr; 15.4 ± 0.5 °C; average depletion = 25% ± 6%)								
NP-1	113 ± 1	119 ± 8 (5)	140 ± 4 (2)	125 ± 7	160 ± 10 (19)	22 ± 6	8 ± 2	24 ± 8
NP-18	81 ± 8	197 ± 3 (4)	229 (1)	203 ± 19	285 ± 15 (14)	29 ± 8	6 ± 2	23 ± 7

*Site descriptions (underlined) include lithology, average precipitation, mean annual temperature, and average chemical depletion fraction (weighted by inverse variance).

†Denudation rates inferred from cosmogenic nuclide concentrations in quartz stream sediment. For methodology see supplemental information (text footnote 1).

§Zr concentrations are measured by X-ray fluorescence and reported as averages ± standard errors; number of samples in parentheses. [Zr]$_{rock}$ is averaged from the combined pool of saprolite and outcrop samples. Our procedures for sampling representative soil and rock are reported in Appendix 1 (see text footnote 1). N.D. = not determined.

**Weathering rates (W) inferred from equation 3. See Appendix 1 (text footnote 1) for details on how we calculate W_{Si}, the silicon chemical weathering rate.

$$W = D \times (1 - [Zr]_{rock}/[Zr]_{soil}). \tag{3}$$

Thus, we can use concentrations of insoluble elements to infer chemical weathering rates from measurements of total denudation rates. The factor $(1 - [Zr]_{rock}/[Zr]_{soil})$ equals the fraction of total denudation that is accounted for by chemical weathering, and is hereafter termed the chemical depletion fraction. Long-term denudation rates of catchments can be measured from cosmogenic nuclide concentrations in sediment collected from catchment streams (e.g., Riebe et al., 2000; also see Appendix 1[1]). According to equation 3, denudation rates measured from cosmogenic nuclides can be used to infer long-term, catchment-wide chemical weathering rates, if representative Zr concentrations for catchment soils and parent rock can be estimated.

Sampling Rationale

We used this new soil mass-balance technique to measure long-term weathering rates at six sites in the Sierra Nevada, California. Our sites are outside the limits of late Pleistocene and Holocene glaciation, are underlain by granitic bedrock, and span a wide range of climates (Table 1), with mean annual temperatures ranging from 4 to 15 °C, and average precipitation from 22 to 145 cm/yr. Hillslopes are soil mantled, with occasional outcrops of boulders and bedrock, and field observations suggest that sediment transport is dominated by diffusive processes such as tree throw, rainsplash, and soil creep. In environments such as these, the assumption that soil depths are in steady state should

be reasonable (see footnote 1 for further discussion). In a separate study, we used cosmogenic nuclides to infer total denudation rates for a series of catchments within each site (Riebe et al., 2000). We found that, within any given site, denudation rates vary by as much as 15 fold across the study catchments. With measurements of chemical weathering rates at each catchment, we can explore how denudation rates affect weathering rates within each site and also how weathering depends on climate. To measure chemical weathering rates, we first estimated catchment-averaged chemical depletion fractions $(1 - [Zr]_{rock}/[Zr]_{soil})$ using average [Zr] from widely distributed rock and soil samples (Table 1). We then used these chemical depletion estimates to infer long-term chemical weathering rates from our cosmogenic measurements of total denudation rates.

RESULTS

At our Fall River and Fort Sage study sites, where denudation rates vary by more than 10 fold, chemical depletion fractions are nearly constant (Fig. 1, A and C). Across a wide range of total denudation rates, the fractional contribution of chemical weathering to denudation is roughly uniform. That these soils are equally depleted by chemical weathering implies that chemical weathering must be faster where physical erosion rates (and thus total denudation rates) are faster (Fig. 1). Chemical depletion fractions inferred from zirconium concentrations are measured independently of denudation rates inferred from cosmogenic nuclides. Therefore, even though chemical weathering rates are calculated directly from total denudation rates (equation 3), the strong coupling between chemical weathering and total denudation (Fig. 1, B and D) is not an artifact of our methods, but instead arises because chemical depletion fractions are roughly constant from catch-
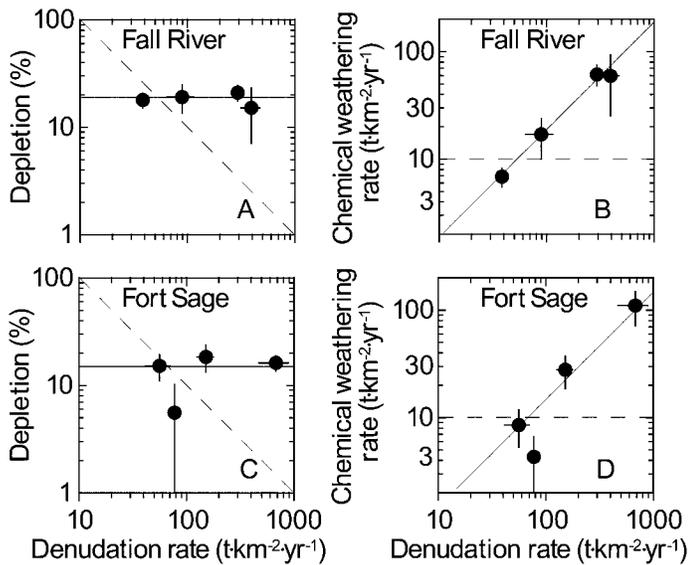
Figure 1. Chemical depletion fractions and chemical weathering rates both plotted against denudation rates for two study sites where denudation rates vary substantially. Chemical depletion fractions are relatively uniform from catchment to catchment at each site, whereas chemical weathering rates increase systematically with denudation rates. Solid lines correspond to relationships based on site-wide averages of chemical depletion fractions (Table 1). Dashed lines correspond to relationships we would have observed if chemical weathering rates were constant from catchment to catchment (as they would have been if they were decoupled from physical erosion rates). In that case, soils at rapidly eroding catchments would have been fresher, as shown in panels A and C. At other four sites (not shown), denudation rates are much less variable (factor of only 1.5; see Table 1), making it difficult to determine how strongly chemical weathering rates are correlated with total denudation rates.
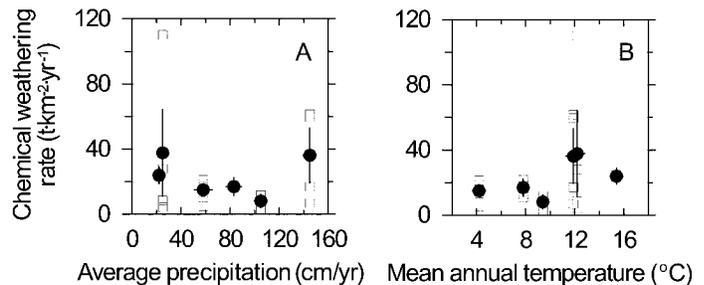

Figure 2. Chemical weathering rates plotted against average annual precipitation (A) and mean annual temperature (B) for individual catchments (open squares) and for site-wide averages (closed circles). Chemical weathering rates show no clear dependence on average precipitation or temperature.

ment to catchment at Fall River and Fort Sage (Fig. 1, A and C). If chemical weathering rates were constant across these sites, faster eroding soils would have been chemically fresher, and our results would have shown chemical depletion fractions decreasing systematically with increasing physical erosion rates (dashed lines of Fig. 1).

DISCUSSION
Possible Mechanisms for Uniform Depletion

We would expect chemical weathering rates to be strongly correlated with physical erosion rates wherever they depend on the rates that fresh rock fragments are supplied to soils (which should be higher in rapidly eroding landscapes). We further expect chemical weathering rates to be coupled with mineral supply rates if chemical depletion makes soils significantly less weatherable, either because the most reactive surfaces are consumed (Lee et al., 1998), or because insoluble mineral coatings precipitate on weatherable surfaces (Nugent et al., 1998). Our Zr measurements are consistent with the hypothesis that chemical weathering rates depend on how quickly rock is incorporated into soil. The average [Zr] in saprolite (i.e., the chemically altered, but physically intact, bedrock at the base of the soil) is roughly the same as average [Zr] in rock outcrops (Table 1; see also footnote 1), indicating that the saprolite has undergone little chemical depletion, even though its appearance and its friability indicate that its mineralogy has been altered. These results suggest that little chemical weathering occurs until rock fragments are incorporated into soil (by processes such as animal burrowing, tree throw, freeze-thaw, and wetting and drying) and thus provide a plausible mechanism for the strong coupling of weathering and erosion shown in Figure 1.

Effects of Climate on Chemical Weathering Rates

Previous work has shown that precipitation and temperature should regulate weathering rates both directly (e.g., White and Blum, 1995) and indirectly (by affecting vegetation, a regulator of weathering; e.g., Moulton and Berner, 1998). If weathering rates were strongly dependent on environmental factors (such as average precipitation and temperature) then, given that each of our Sierran sites represents a distinct, roughly uniform climatic regime, we would expect weathering rates to be roughly consistent from catchment to catchment at each site. In that case, chemical depletion fractions would vary inversely with denudation rates, in contrast to what we observe in Figure 1. Uniform chemical depletion across our catchments indicates that differences in chemical weathering rates are strongly associated with differences in physical erosion rates. This is one reason why chemical weathering rates show no clear correlation with either mean annual temperature or average annual precipitation (Fig. 2); any effects of climate on chemical weathering rates may be obscured by the large variations in physical erosion rates among the catchments at each individual site.

Even when the effects of erosion rates are accounted for in multiple regression analyses, the correlations between chemical weathering rates and precipitation and temperature are not statistically significant. Multiple regression shows that chemical weathering rates increase at 0.16 ± 0.01 t·km^{-2}·yr^{-1} for each 1 t·km^{-2}·yr^{-1} increase in total denudation rate (significance level <0.0001), 0.64 ± 0.39 t·km^{-2}·yr^{-1} for each 1 °C rise in temperature (significance level >0.10), and 0.50 ± 3.10 t·km^{-2}·yr^{-1} for each 1 m/yr of precipitation (significance level >0.85). The lack of correlation between chemical weathering rates and climate persists even when the rapid chemical weathering rates of the quickly eroding sites at Fall River and Fort Sage are excluded from the analysis. Thus, we infer that across the wide range of temperature and precipitation regimes represented by our sites, any effects of climate on weathering rates are small compared to the effects of erosion rates.

The mass-balance approach averages chemical weathering rates over thousands of years, whereas instrument records of climate span years to decades. Our analysis could be confounded if climate has differed significantly over the two time scales, but paleoclimate studies in the Sierra Nevada region suggest that, over the past several thousand years, average temperature and precipitation have changed little, at least compared to the differences in climate among our study sites. For example, tree-ring records and tree-line reconstructions from bristlecone pine show that, over the past 5500 yr, temperatures have varied by <2 °C in the nearby White Mountains (LaMarche, 1974). Furthermore, paleosalinity records from San Francisco Bay sediments indicate no overall trend in Sierran river discharge over the past 2700 yr (Ingram et al., 1996). Moreover, late Holocene hydrologic fluctuations

were largely synchronous across the western United States (Earle, 1993), implying that they affected the Sierra Nevada as a whole, and would not have substantially altered the site-to-site climatic differences on which our analysis is based.

Climatic effects on weathering rates may be easier to distinguish across sites with similar erosion rates. For example, the short-term silicate weathering flux for granitic Rio Icacos, Puerto Rico, has been estimated to be between 8 and 23 t·km^{-2}·yr^{-1} (McDowell and Asbury, 1994; White et al., 1998), roughly 3 times higher than silicate weathering rates (range = 2–8 t·km^{-2}·yr^{-1}) of Sierran catchments with denudation rates ~100 t·km^{-2}·yr^{-1} (approximately the denudation rate at Rio Icacos, based on cosmogenic nuclide measurements; Brown et al., 1995). Weathering rates are relatively high at Rio Icacos, not because its lithology is different or because its erosion rates are higher, but apparently because its soils can be more intensely weathered (for a given erosion rate) in the hotter, wetter climate (mean annual temperature = 22 °C, and average precipitation = 420 cm/yr). The chemical depletion fraction of Rio Icacos is ~63% (estimated from [Zr] in ridge-top soils and bedrock; White et al., 1998), significantly higher than chemical depletion fractions of our Sierran sites (maximum = 29%). Thus, chemical depletion fractions may vary systematically with climate, even though chemical weathering rates do not (due to the overwhelming effects of differences in mineral supply rates from erosion).

The silicate weathering rate of tropical Rio Icacos, 9–23 t·km^{-2}·yr^{-1}, was the most rapid granitic catchment weathering rate reported to date (White et al., 1998), but is equaled and surpassed by those of the rapidly eroding, temperate Sierran catchments (19–36 t·km^{-2}·yr^{-1} for our catchments FR-2, FR-5, and A4(s); see Table 1). Faster weathering rates in these Sierran catchments are explained by their total denudation rates being three to seven times faster than Rio Icacos (~300–700 versus 100 t·km^{-2}·yr^{-1}), while their chemical depletion fractions are only two to three times smaller. These observations highlight the potential for strong tectonic control on chemical weathering rates. Denudation rates in soil-mantled terrain can vary by orders of magnitude with tectonic setting (Milliman and Syvitski, 1992; Riebe et al., 2000). However, chemical depletion fractions, which apparently depend on climate, vary by only about a factor of six in granitic terrain (~10% at Sunday Peak to ~60% at Rio Icacos), at least across the range of climates considered here.

IMPLICATIONS

The hypothesis that physical erosion and chemical weathering are interrelated (Stallard and Edmund, 1983) has not previously been tested with measurements of long-term rates of chemical weathering and physical erosion. Our measurements confirm that, over time scales of soil formation and landscape evolution, rates of physical erosion and chemical weathering are tightly coupled.

Our results also demonstrate that erosional effects on weathering rates are significantly larger than any climatic effects across our sites. The lack of correlation between chemical weathering and climate implies that long-term climate shifts will not substantially dampen or amplify themselves through feedbacks with chemical weathering rates on soil-mantled hillslopes, unless climate changes are great enough to significantly affect the chemical depletion of soils. Our results do not rule out the potential for climate feedbacks in lowlands and flood plains, if their chemical weathering rates are less sensitive to mineral supply rates, and thus relatively more sensitive to climatic factors. The strong correlation between erosion rates and weathering rates measured here confirms earlier suggestions (Raymo et al., 1988) that shifts in mountain uplift rates, and thus erosion rates, will lead to changes in weathering rates, which may affect atmospheric CO_2 concentrations and alter climate over long time scales.

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