Influence of late Cenozoic mountain building on ocean geochemical cycles

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ABSTRACT

In a steady-state ocean, input fluxes of dissolved salts to the sea must be balanced in mass and isotopic value by output fluxes. For the elements strontium, calcium, and carbon, rivers provide the primary input, whereas marine biogenic sedimentation dominates removal. Dissolved fluxes in rivers are related to rates of continental weathering, which in turn are strongly dependent on rates of uplift. The largest dissolved fluxes today arise in the Himalayan and Andean mountain ranges and the Tibetan Plateau. During the past 5 m.y., uplift rates in these areas have increased significantly; this suggests that weathering rates and river fluxes may have increased also. The oceanic records of carbonate sedimentation, level of the calcite compensation depth, and δ13C and δ87Sr in biogenic sediments are consistent with a global increase in river fluxes since the late Miocene. The cooling of global climate over the past few million years may be linked to a decrease in atmospheric CO2 driven by enhanced continental weathering in these tectonically active regions.

INTRODUCTION

Changes in sea-floor-spread rates, land-sea distribution, and rates of outgassing have been proposed as primary causes of the decline in global temperatures over the past 100 m.y. (e.g., Hays and Pitman, 1973; Berner et al., 1983). On shorter time scales, tectonic changes in oceanic sills depths (Keigwin, 1982), increased volcanism (Kennett and Thunell, 1975), and epeirogenic uplift in highland regions of northern Canada (Birchfield et al., 1982) have been invoked as possible causes of the late Pliocene appearance of continental ice sheets in the Northern Hemisphere. It has been proposed that rapid uplift of the Himalayan-Tibetan Plateau region and the plateau and mountain regions of the southwest United States could also have caused climates to cool during the past 3 m.y. (Ruddiman and Raymo, 1988). Increased elevation in these areas may have enhanced the meridionality of Northern Hemisphere atmospheric waves and allowed greater southward penetration of polar air masses over east-central North America and Europe.

Here, we propose that increasing rates of uplift and erosion in the Himalayas, Andes, and Tibetan Plateau have contributed to a significant increase in the global rate of chemical weathering since the late Miocene. We focus on the Himalayan-Tibetan region and the Andes because their associated rivers carry the greatest dissolved and particulate loads to the sea, and thus large changes in the amount of erosion in these areas would have the greatest global effect. However, we note that rates of uplift have increased during the late Neogene for many other areas, including the southwest United States, the Swiss Alps, and the Southern Alps of New Zealand.

Using simple one-box ocean models of the geochemical cycles of strontium (Sr), calcium (Ca), and carbon (C), we show that the marine records of carbonate sedimentation, calcite compensation depth (CCD), δ87Sr, and δ13C are consistent with increased fluxes of weathering products to the sea over the past 5 m.y. We propose that this increase in the rate of global chemical weathering could have lowered atmospheric CO2 concentrations. This view differs from recent geochronological models that ignore relief as a factor controlling global weathering rates and atmospheric CO2 (e.g., Berner et al., 1983; Volk, 1987).

EVIDENCE FOR PLEISTOCENE: PLEISTOCENE MOUNTAIN UPLIFT

Mineralogic and biologic evidence suggests that rates of uplift and erosion have increased substantially in the Himalayas since the early Pliocene. Rb/Sr, K/Ar, and fission-track ages of minerals in the Siwalik formation and other molasse deposits along the southern margin of the Himalayan range indicate that progressively faster rates of uplift, erosion, and sedimentation have occurred during this interval (e.g., Saini et al., 1979; Burbank and Johnson, 1983). These formations coarsen progressively upward from early Pliocene silts to middle and late Pleistocene conglomerates, consistent with increasing uplift through the past 5 m.y. Mammalian distributions (the Hipparion fauna) also suggest that the Himalayas were not an effective barrier to north-south migration until the late Pliocene (West, 1984).

On the Tibetan Plateau, late Miocene and early Pliocene subtropical pollen assemblages, indicative of elevations below 1.5 km, are found today at elevations higher than 4 km (Hsu, 1978). Results from 12 additional pollen and vegetation studies spanning the Cenozoic are summarized in Figure 1a. Over the past 5 m.y., uplift rates have been at least twice as great (curves 2N and 2S) as those of the previous 5 m.y., and may have increased more than fivefold (curve 1).

In the eastern Cordillera and Altiplano of the Bolivian Andes, Benjamin et al. (1987) used dated pairs of apatite and zircon to estimate Neogene uplift rates. Their results (Fig. 1b) showed that uplift rates have approximately doubled over the past 5 m.y. To the south, the Puna Plateau of Argentina also underwent a major phase of uplift, beginning between 10 and 5 Ma, which culminated at approximately 2 Ma (Allmendinger, 1986). To the north, in western Colombia, the timing of major uplift centered on 3 to 4 Ma (McCourt et al., 1984). As in the Tibetan-Himalayan region, uplift rates appear to have increased significantly since the late Miocene, even though the exact timing and magnitude of these changes are still poorly constrained.

UPLIFT AND CHEMICAL WEATHERING RATES

Increased rates of vertical movement in the Himalayas, Tibet, and the Andes could have a significant effect on chemical weathering rates and the global flux of dissolved material to the sea. Today, the three largest sources of dissolved materials are the Yangtze, Amazon, and Ganges-Brahmaputra rivers (Berner and Berner, 1987). These three rivers, which drain the Tibetan Plateau, Andes, and Himalayas, respectively, account for about 20% of the total dissolved plus suspended solids and water discharges to the ocean. Including all the other rivers draining these three areas, chemical erosion in the Himalayas, Andes, and Tibet makes a substantial contribution to the flux of dissolved material reaching the sea.

Relief influences rates of chemical weathering and dissolved fluxes to the sea for three reasons:

1. Rapid mechanical breakdown of rocks in areas of rapid uplift enhances exposure of primary minerals to chemical weathering (Stallard and Edmond, 1983). Abrasion in mountain streams also removes iron-oxide coatings that inhibit chemical attack and retard weathering.

2. Dissolved loads are highest in areas dominated by easily eroded sedimentary rocks. In general, areas of active tectonism are character-
Figure 1. a: Estimated elevation of Tibet over past 60 m.y. based on 12 vegetation studies (after Mercier et al., 1987). Curve 1 shows maximum rates of uplift since Miocene; curves 2S and 2N show minimum uplift rates for southern and northern Tibetan Plateau, respectively. b: Mineralogic estimates of uplift in Peruvian Altiplano (after Benjamin et al., 1987) show that uplift rates have more than doubled in this area over past 5 m.y. c: Global CaCO3 sedimentation rates (after Delaney and Boyle, 1988). d: Average marine δ87Sr values plotted at 2 m.y. increments over past 60 m.y.; arrow shows steady-state value for current measured Sr inputs (data from Elderfield, 1986). e: Average global calcite compensation depth (CCD) over Cenozoic (after Delaney and Boyle, 1988). f: δ13C of bulk carbonate recovered from Deep Sea Drilling Project Sites 525 and 528, South Atlantic (Shackleton, 1987).
ized by uplifted sedimentary formations typical of continental margins, whereas thick, well-weathered soils and more resistant crystalline rocks characterize peneploplained, continental lowlands (Stallard and Edmond, 1983). (3) Chemical and physical weathering rates increase with runoff, which tends to be greatest in mountainous areas because of the orographic influence of mountain ranges on precipitation patterns. Schmitz (1987) estimated that the annual rainfall associated with the Asian monsoon may have increased as much as tenfold in conjunction with Himalayan uplift.

The above effects have been well documented in the Amazon River system, where over 75% of the discharged dissolved solids are eroded from the Andean highlands (Gibbs, 1967; Stallard and Edmond, 1983). Thus, increasing rates of uplift in the Himalayan-Tibetan and Andean regions could have significantly enhanced dissolved fluxes in rivers to the ocean since the late Miocene and early Pliocene. Are sea-floor records of $^{87}$Sr, carbonate sedimentation, CDD, and $^{813}$C consistent with such an increase?

**RIVER FLUXES AND OCEAN GEOCHEMICAL CYCLES**

We use steady-state, one-box models of the ocean to estimate the direction and approximate magnitude of changes in river fluxes needed to explain the evolution of the $^{87}$Sr and carbonate accumulation output records over the past 5 m.y. The record of $^{813}$C cannot be used to constrain estimates of past river fluxes for reasons discussed below. Mass-balance equations for the oceanic constituents Sr, Ca, and C are given in Appendix 1. Over thousands of years, the ocean achieves a steady-state mass balance by adjusting outputs so that they equal inputs. Potential oceanic feedback mechanisms that govern changes in output rates have been discussed by Broecker and Peng (1982).

The modern input and output fluxes, as well as isotopic values of the Sr and C fluxes, are given in Table 1. The flux and isotopic values of river Sr are based on recent measurements of rivers draining the Himalayan and Andean regions (Edmond, 1987, and unpub. data). These values are higher than those measured in previous studies of limited areal extent (e.g., Martin and Meybeck, 1979; Wadleigh et al., 1985) and are consistent with the hydrothermal fluxes based on $^3$He loss from the ocean crust (Edmond, 1987). The Sr and Ca output fluxes are calculated from measured input fluxes, and C fluxes are based on a 4:1 burial ratio of C in carbonate and organic matter (Broecker and Peng, 1982), with the additional constraint that the output fluxes of C and Ca in carbonate sediments must be equal.

In Table 1, our isotopic value of Sr in modern carbonate (and hence seawater) is not the current measured seawater value ($^{87}$Sr = 0.07‰; Palmier and Elderfield, 1985). The oceanic isotopic value for Sr cannot be at steady state because the residence time of Sr in the ocean is ~5 m.y. (Broecker and Peng, 1982) and the oceanic record of Sr isotopes (Fig. 1d) shows a continual increase over the past 5 m.y. (indicating that inputs and/or outputs are changing). If the ocean were allowed to reach steady state with the measured river, redissolution, and hydrothermal inputs (Table 1), we calculate that the seawater-carbonate $^{87}$Sr value would be about 1.01‰. We adopt this value to represent the modern ocean. The isotopic value for river carbon ($^{813}$C = 39‰) was determined from the isotopic value of bulk marine carbonate ($^{813}$C = +0.3‰; Shackleton, 1987) and deep-sea sediment organic carbon ($^{813}$C = -21‰; Degens, 1969) and from the fractions of each being deposited in the modern ocean (Broecker and Peng, 1982).

For each element in Table 1, rivers dominate the inputs to the ocean reservoir. Hydrothermal and redissolution inputs are less significant. Output fluxes occur primarily by biogenic sedimentation of carbonates. To avoid the possibility of large variations in spreading rates and hydrothermal fluxes (which we assume remain constant), we restrict this study to the past 5 m.y., and we specifically address the influence of rapid vertical tectonics on erosional fluxes. Output fluxes for the 5 Ma ocean (Table 1) are estimated below.

**Strontium**

The $^{87}$Sr record of oceanic carbonate is shown in Figure 1d. Unlike the present-day value, the 5 Ma Sr isotopic value ($^{87}$Sr = 0.28‰) is assumed to be near steady state because of the relative constancy of $^{87}$Sr values over the previous 10 m.y. (15-5 Ma). This implies an early Pliocene river Sr flux of $1.85 \times 10^{10}$ mol/yr. We assume no change with time in the isotopic value of the river input and, thus, in the proportion of silicates to carbonates being weathered on the continents. If the fraction of carbonates being weathered has increased since the early Pliocene (as might be expected from the geology of Tibet, the Andes, and the Himalayas), then our 5 Ma river Sr flux would be slightly overestimated. Holding all else constant, the Sr isotopic values indicate that the river flux of Sr has increased by about 55% since the early Pliocene.

**Calcium**

Because we do not know the fraction of biogenic aragonite deposited in the past or even its bulk average Sr/Ca ratio, we cannot use the Sr/Ca ratio in foraminifera to estimate past river fluxes of Ca. The global record of carbonate accumulation, however, provides some constraints on estimates of dissolved river inputs.

Davies et al. (1977) estimated average CaCO$_3$ sedimentation rates in the world’s oceans over the Cenozoic by using core data recovered by Legs 1-34 of the Deep Sea Drilling Project (DSDP). Their compilation, with the revised time scale used by Delaney and Boyle (1968), is plotted in Figure 1c. Davies et al. (1977) suggested that increases in CaCO$_3$ sedimentation rates were caused by greater river delivery of Ca. Taken at face value, the trends in Figure 1c would indicate that global CaCO$_3$ sedimentation rates have approximately doubled since the early Pliocene, and therefore, the output flux of Ca in CaCO$_3$ sediments has also doubled. This would imply that river erosional fluxes of Ca have increased by as much as 160% (using mass-balance equations in App. 1).

The true increase, however, is probably smaller than is suggested by Figure 1c. Moore and Heath (1977) argued that the late Cenozoic increase in CaCO$_3$ sedimentation was an artifact of enhanced preservation of younger sediments and not due to increased river fluxes. Accept-

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**TABLE 1. MASS BALANCE FLUX ESTIMATES**

<table>
<thead>
<tr>
<th></th>
<th>Sr (10$^{12}$mol/yr)</th>
<th>$^{87}$Sr (‰)</th>
<th>Ca (10$^{12}$mol/yr)</th>
<th>C (10$^{12}$mol/yr)</th>
<th>$^{813}$C (‰)</th>
</tr>
</thead>
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<tr>
<td>Modern Oceans</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>River input</td>
<td>2.9(a)</td>
<td>5.36(a)</td>
<td>13(e)</td>
<td>20</td>
<td>-3.9</td>
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<tr>
<td>Hydrothermal input</td>
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<td>-8.74(d)</td>
<td>30</td>
<td>-</td>
<td></td>
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<tr>
<td>Redissolution input</td>
<td>0.5(c)</td>
<td>-0.85(c)</td>
<td>-</td>
<td>4.0</td>
<td>-</td>
</tr>
<tr>
<td>Hydrothermal output</td>
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<td>+1.01</td>
<td>12</td>
<td>4.0</td>
<td>-</td>
</tr>
<tr>
<td>Organic output</td>
<td>-</td>
<td>-</td>
<td>16</td>
<td>16</td>
<td>+0.38(i)</td>
</tr>
<tr>
<td>Carbonate output</td>
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<td>+1.01</td>
<td>-</td>
<td>16</td>
<td>-</td>
</tr>
<tr>
<td>5 Ma Oceans</td>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>River input</td>
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<td>5.36*</td>
<td>9</td>
<td>16.2</td>
<td>-3.9*</td>
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<tr>
<td>Hydrothermal input</td>
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<td>-8.74*</td>
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<td>-</td>
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<tr>
<td>Redissolution input</td>
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<td>-0.85*</td>
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<tr>
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<td>-</td>
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</tr>
<tr>
<td>Organic output</td>
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<td>-</td>
<td>4.2</td>
<td>-19.8</td>
<td>-</td>
</tr>
<tr>
<td>Carbonate output</td>
<td>2.35</td>
<td>-0.28(j)</td>
<td>12(k)</td>
<td>12</td>
<td>+1.6(i)</td>
</tr>
</tbody>
</table>

Note: Letters with values indicate references: a-Edmond (unpub. data and 1987); b-Palmer and Elderfield (1985); c-Elderfield and Gieskes (1982); d-Albarde et al. (1981); e-Martin and Meybeck (1979); f-Von Damm et al. (1985); g-Broecker and Peng (1982); h-Degens (1969); i-Shackleton (1987); j-Elderfield (1986); k-Davies et al. (1977), with timescale corrections of Delaney and Boyle (1988) assuming 50% preservation effects and 50% true changes in sedimentation rates. Values with no reference were determined by mass-balance difference. * Values assumed to be equal to modern.
ing this model of age-dependent preservation, Davies and Worsley (1981) recompiled the carbonate data and found that the increase in sedimentation rates since the Miocene was greatly reduced. Despite the preservation effect, several arguments suggest that a late Cenozoic increase in carbonate deposition is plausible. First, dissolution of CaCO\(_3\) and hence destruction of sediment, can occur either shortly after deposition (below the CCD) or much later, long after burial (by a shaling of the CCD). The fact that the CCD has deepened by ~350 m since the early Pliocene (Fig. 1e) suggests that dissolution may have been more widespread at 5 Ma than today. Increased preservation of CaCO\(_3\) over this interval would then be not solely a function of age but a consequence of the CCD deepening, possibly in response to increased river fluxes of Ca.

Second, in their evaluation of preservation effects, Moore and Heath (1977) defined a hiatus as any DSDP section that accumulated at rates less than 1 m/my. If, on average, sedimentation rates were much lower in the past, as suggested by Figure 1c, then sediments accumulating at less than 1 m/my would greatly expand in areal coverage. This would increase the frequency of sedimentary sequences assigned as hiatuses in the Moore and Heath analysis and create an artificial preservation effect.

Thus, it is unclear what fraction of the increase in carbonate sedimentation is due to increased river inputs and what fraction is an artifact of age and preservation. We suggest that Figure 1c represents the maximum possible increase in global carbonate sedimentation over that past 5 my.; the true increase is probably lower because of the preservation effect discussed by Moore and Heath (1977). In Table 1, we ascribe 50% of the apparent increase in Figure 1c to preservation and 50% to a true increase in carbonate sedimentation rates. This model implies that river Ca fluxes have increased by ~45% since the early Pliocene. This estimate is more in keeping with the increase in river fluxes suggested by the Sr data, but larger (or smaller) increases cannot be ruled out. If, indeed, carbonate burial rates have increased since the early Pliocene and the level of the CCD has dropped from 4150 to 4500 m (Fig. 1e), then the CaCO\(_3\) production rate would have increased from 25 \times 10\(^{12}\) mol/yr to 28 \times 10\(^{12}\) mol/yr (calculation in App. 2). Thus, enhanced oceanic carbonate productivity would also have accompanied an increase in dissolved fluxes in rivers.

**Carbon**

Because equal amounts of Ca and C are buried with CaCO\(_3\), the output flux of C in carbonates will equal the output flux of Ca in carbonate sediments at 5 Ma. But to independently constrain the river flux of C at this time also requires calculation of the burial of organic matter from two of the following three parameters: (1) the burial ratio of carbonate to organic matter in the ocean; (2) the \(\delta^{13}C\) of the buried bulk-marine organic matter; (3) the \(\delta^{13}C\) of the buried bulk-marine carbonate. Unfortunately, only the bulk carbonate \(\delta^{13}C\) record is known in sufficient detail.

The \(\delta^{13}C\) of bulk carbonate from DSDP Sites 525 and 528 is shown in Figure 1f. Parts of this record have been reproduced in several areas (Shackleton, 1987), suggesting that it is a representative global signal. Likewise, the present-day value (\(\delta^{13}C = +0.38 \pm 0.28 \%_{oo}\); Shackleton, 1987) is found over a wide range of latitudes in three oceans, suggesting that the decline in bulk carbonate \(\delta^{13}C\) from the Miocene to the present is also global.

Shackleton (1987) estimated burial rates of marine organic matter through the Cenozoic from his bulk carbonate \(\delta^{13}C\) record alone (Fig. 1f) by assuming that (1) the carbon isotope fractionation \(\delta^{13}C_{org} - \delta^{13}C_{calc}\) has remained constant, (2) the burial flux of CaCO\(_3\) was constant, and (3) the \(\delta^{13}C\) of the river input was constant. His assumptions led directly to his conclusion that \(\delta^{13}C_{org}\) burial, organic carbon/carbonate burial ratios, and river-C inputs decreased toward the present.

Here, we recalculate \(\delta^{13}C_{org}\) burial at 5 Ma, also using Shackleton's bulk carbonate \(\delta^{13}C\) record (Fig. 1f). Instead of assuming a constant burial flux of CaCO\(_3\), however, we use the lower carbonate output flux at 5 Ma (12 \times 10\(^{12}\) mol/yr; Table 1) derived in the previous section. By using \(\delta^{13}C_{calc} = +1.6 \%_{oo}\) at 5 Ma (from Fig. 1f), we estimate the \(\delta^{13}C_{org}\) sedimentation flux in the early Pliocene to be 4.2 \times 10\(^{12}\) mol/yr (vs. 4 \times 10\(^{12}\) mol/yr today), and the river-C flux to be 16.2 \times 10\(^{12}\) mol/yr (vs. 20 \times 10\(^{12}\) mol/yr today). Both Shackleton's and our estimates imply that a greater fraction of C was deposited in organic matter at 5 Ma and that total organic carbon deposition was also greater (although our estimate of the latter difference is much less than Shackleton's). However, the calculated river fluxes differ significantly. When we allow carbonate sedimentation rates to increase toward the present, river-C fluxes also increase (in the model presented here, by about 25%), whereas in Shackleton's model, which assumes constant carbonate sedimentation rates, river-C fluxes decrease toward the present.

One potential problem with both treatments of the C cycle is the assumption that \(\delta^{13}C\) fractionation factors remain constant. Measurements of \(\delta^{13}C\) within single species of planktonic foraminifers suggest that surface-water \(\delta^{13}C\) has not changed significantly since the late Miocene (Savin et al., 1985). Acceptance of these measurements and the record of bulk carbonate \(\delta^{13}C\) implies that biological fractionation effects must have changed. If so, then it is no longer possible to estimate the relative proportions of carbonate and organic sedimentation and hence past river fluxes of C, without data for \(\delta^{13}C\) of bulk organic matter. If, rather than assuming constant fractionation factors, one assumes a constant organic carbon/carbonate output ratio, then the organic matter and river-C fluxes both must increase by 33% toward the present, as prescribed by the change in the carbonate flux. This would imply that fractionation factors were larger in the early Pliocene ocean, possibly because of elevated CO\(_2\) concentrations in the ocean (Arthur et al., 1985).

**DISCUSSION**

The Sr isotope record and carbonate sedimentation curve are both consistent with an increase in dissolved fluxes in rivers over the past 5 my. The bulk carbonate \(\delta^{13}C\) record is also consistent with increased river fluxes, provided the model invoked to explain this record does not assume constant burial fluxes a priori. Such an increase in the rate of global chemical erosion could have been caused by increased rates of tectonic uplift in the Himalayas and Andes and on the Tibetan Plateau, as well as in other active tectonic regions such as the western United States and the Alps. However, alternative mechanisms could have produced the observed \(\delta^{87}Sr\) and carbonate records. The increase in the \(\delta^{87}Sr/\delta^{86}Sr\) ratio could have been caused by a 40% decrease in hydrothermal fluxes over this interval, or by a 95% decrease in the redissolution flux. Increasing the proportion of silicates weathered on land by ~16% could also explain the evolution of Sr isotopes since the late Miocene (Palmer and Elderfield, 1985). Yet all these explanations would require constant or decreasing Ca-input fluxes, opposite to the trend suggested by both the CaCO\(_3\) sedimentation and CCD records.

The apparent increase in global erosional fluxes has also been attributed to the growth of continental ice sheets and their effect on erosion, both directly through the exposure and grinding of rock (Armstrong, 1971) and indirectly through lowering sea level and increasing land area subject to erosion (DePaolo, 1986). Both mechanisms have probably influenced fluxes to the sea, although the relative importance of these sources compared to low-latitude rivers (that dominate the dissolved chemical influx today) remains uncertain.

The increase in chemical weathering that we ascribe to uplift may have influenced atmospheric CO\(_2\) during weathering reactions or by influencing the oceanic carbon cycle. The increased supply of cations associated with enhanced continental erosion would tend to drive ocean alkalinity up and CO\(_2\) down (Broecker and Peng, 1982). Indeed, if relief is an important factor in continental weathering, as we argue here, then CO\(_2\)
levels may be controlled not only by the balance between sea-floor spreading rates, land-sea distributions, and atmospheric CO₂ feedbacks on weathering rates (e.g., Berner et al., 1983; Volk, 1987) but also by relative rates of sea-floor spreading (CO₂ production) and continental weathering (CO₂ consumption).

It is likely, the onset of Northern Hemisphere glaciation may have been related to mountain uplift through the impact of high plateaus and mountain ranges on atmospheric circulation patterns (Ruddiman and Raymo, 1988) and the impact of enhanced rates of chemical erosion on the oceanic C cycle and atmospheric CO₂. At present, we cannot adequately quantify how changes in weathering rates, silicate/carbonate weathering ratios, carbonate/organic carbon burial ratios, and ocean ventilation rates might alter Earth’s CO₂ budget. Nevertheless, uplift and erosion of mountain ranges can clearly influence chemical fluxes through the oceans and atmosphere, and thus, on climate, as scales of 10⁶–10⁷ yr.

APPENDIX 1. STEADY-STATE MASS-BALANCE EQUATIONS

\[ \begin{align*}
\text{Ca} & \quad \text{Ca}_{\text{in}} - \text{Ca}_{\text{out}} = \text{Ca}_{\text{dep}} - \text{Ca}_{\text{erod}} \\
\text{Sr} & \quad \text{Sr}_{\text{in}} - \text{Sr}_{\text{out}} = \text{Sr}_{\text{dep}} - \text{Sr}_{\text{erod}} \\
\text{Fe}_{\text{carb}} & \quad \text{Fe}_{\text{in}} - \text{Fe}_{\text{out}} = \text{Fe}_{\text{dep}} - \text{Fe}_{\text{erod}} \\
\text{Al}_{\text{carb}} & \quad \text{Al}_{\text{in}} - \text{Al}_{\text{out}} = \text{Al}_{\text{dep}} - \text{Al}_{\text{erod}}
\end{align*} \]

where \( \delta \) is isotopic enrichment/depletion compared to modern seawater (per mil), \( F \) is fluxes to/from ocean, \( r \) is river, \( hyd \) is hydrothermal, \( org \) is organic matter, \( carb \) is carbonate, \( rediss \) is redissolution of carbonates, and \( oc \) is ocean reservoir.

APPENDIX 2. CALCULATION OF SHELL PRODUCTION RATES

Shell production rate = \( \text{F}_{\text{calc}} \times \frac{\text{fractional area above calcite compensation depth (CCD)}}{\text{CCD}} \times \frac{\text{Ca}_{\text{in}}}{16 \times 10^{12} \text{mol/yr (present)}} \)


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