

The Himalayas, organic carbon burial, and climate in the Miocene

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Abstract. Cooling ages of rock in the Himalayas imply that rapid exhumation between the Main Central thrust system and the South Tibetan detachment system occurred between 21 and 17 Ma. The generation of relief and enhanced weathering which followed this event may have resulted in a pronounced increase in the delivery of dissolved strontium, carbon, phosphorus, and other chemical weathering products to the ocean (Richter et al., 1992). The increased supply of nutrients stimulated productivity in oceanic upwelling zones and expansion of the oxygen minimum zone leading to enhanced burial and preservation of organic matter in the Monterey formation and other deposits from this interval. A drawdown of atmospheric CO₂ associated with enhanced chemical weathering rates and organic matter burial may have led to global cooling and the expansion of the Antarctic ice sheet by 15 Ma. The above scenario differs from the "Monterey hypothesis" of Vincent and Berger in that CO₂ drawdown is primarily via silicate weathering rather than organic carbon burial and that organic carbon burial is driven by increased delivery of nutrients to the ocean rather than by stronger upwelling. A carbon mass balance calculation which assumes that river fluxes have been increasing over the last 40 Ma predicts that absolute organic carbon burial increased over this interval while, at the same time, the fraction of carbon buried as organic matter versus carbonate decreased. This implies that the organic carbon cycle has acted as a net source of CO₂ to the atmosphere over the late Cenozoic.

Introduction

Many lines of evidence suggest that global silicate weathering rates have increased significantly over the Cenozoic (see review by Raymo and Ruddiman [1992]). Much of this increase may be attributed to enhanced weathering in the Himalayas, a region of pronounced tectonic activity and monsoon precipitation. Because silicate weathering consumes atmospheric CO₂, it has also been proposed that enhanced weathering over the Cenozoic resulted in global cooling as well [Raymo et al., 1988; Raymo, 1991]. However, a problem with this hypothesis has always been the relatively small size of the atmospheric CO₂ reservoir compared to the rock reservoir. If weathering rates increase slightly while mantle degassing rates remain constant, atmospheric CO₂ levels fall to zero within a few million years [e.g., Sundquist, 1991]. This is why almost all numerical carbon cycle models have weathering rates controlled by mean global temperature [e.g., Berner et al., 1983; Berner, 1990]. This way, as climate cools, chemical weathering rates decrease, keeping the climate system and atmospheric CO₂ levels in

check. However, these models are at odds with strontium isotopic evidence for increasing weathering over the last 40 m.y. [Raymo et al., 1988; Hodell et al., 1990; Raymo, 1991; Richter et al., 1992], a time of pronounced global cooling, as well as with modern river studies [e.g., Edmond, 1992; Krishnaswami et al., 1992]. A number of solutions to this dilemma have been proposed [e.g., Francois and Walker, 1992; Caldiera, 1992] and here I examine the suggestion that changes in the net global organic carbon burial rate act as a negative feedback preventing a runaway icehouse [Raymo and Ruddiman, 1992]. In particular, data from the Miocene (24-5 Ma) show that major changes in tectonics, climate, and organic carbon burial are tightly coupled on timescales of a few million years.

Himalayan Weathering

A major phase of deformation in the Himalayan orogen occurred between 21 and 17 m.y. ago. Rapid mineral cooling rates imply that regional exhumation rates, and probably relief, increased significantly at this time (see recent review by Sorkhabi and Stump [1993]). Hodges et al. [1992] estimate that a minimum of 34 km of displacement and 6 km of unroofing occurred within this 4 m.y. in the Qomolangma belt of the Greater Himalaya,

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Paper number 94PA00289.
0883-8305/94/94PA-00289\$10.00

while *Copeland et al.* [1987] and *Richter et al.* [1992] show evidence for exceptionally high unroofing rates of the Gandese batholith of southern Tibet within this interval. Movement along the Himalayan Main Central thrust system (MCTS) was also greatest at this time [*Harrison et al.*, 1992; *Gansser*, 1981; *LeFort*, 1989].

An associated increase in chemical weathering and river nutrient fluxes is suggested by the strontium isotopic curve of seawater which shows a rapid increase to more radiogenic values at this time (Figure 1a). (Note that the stratigraphic age of the positive inflection in $\delta^{87}\text{Sr}$ is controversial, with estimates ranging from 22.8 Ma [*Oslick et al.*, this issue] to 19.7 Ma [*Hodell and Woodruff*, this issue] to 17.9 Ma [*DePaolo and Finger*, 1991].) I agree with *Richter et al.*'s [1992] argument that the rapid increase in $\delta^{87}\text{Sr}$ after 20 Ma was related to a major episode of erosion and weathering that occurred in the Himalayan-Tibetan region at this time. While they emphasize erosion of the southern Tibetan Plateau region, it is probable that exhumation and weathering of the high Himalayan crystalline series between the MCTS and the South Tibetan detachment system was the greater source of the strontium to the ocean at this time (C. Burchfiel, personal communication, 1992).

The Monterey Formation

Stratigraphic studies show that immediately following the inflection in the oceanic strontium isotope record, deposition of the Monterey formation began [*DePaolo and Finger*, 1991]. The widespread diatomaceous rocks of the Miocene Monterey formation were deposited around the rim of the Pacific Ocean (and in other oceans as well, although they are less well documented); they generally consists of three units whose boundaries are often indistinct and time transgressive but are typified by 1) a lower to middle Miocene calcareous facies with abundant mudstones and shales, (2) a middle Miocene phosphatic facies, and (3) a middle Miocene to lower Pliocene siliceous facies [*Isaacs et al.*, 1983; *Garrison et al.*, 1981; *Compton et al.*, 1990]. The upper lower Miocene to lower middle Miocene units in particular are characterized by extremely high organic carbon contents, sometimes 25% by weight, and are the major source rock for petroleum off California today (20 billion barrels to date [*Isaacs and Petersen*, 1987]). Likewise, the middle Miocene phosphorite facies, most abundant between 15.5 and 13.2 Ma, represent one of the most significant deposits of this type in the Cenozoic [*Garrison et al.*, 1990; *Compton et al.*, 1990]. Quantitative estimates of P and organic C accumulation rates are difficult to make, since deposits are hard to date and are often time transgressive. Some investigators have the highest organic matter (OM) accumulation rates in the early Miocene (older than 17 Ma [*Isaacs and Petersen*, 1987]), while others have it closer to 15 Ma during the peak of the phosphorus unit [*Pisciotta and Garrison*, 1981].

It has long been recognized that the enhanced removal of OM in the Monterey formation could have resulted in a drop in atmospheric CO_2 and global cooling, and a number

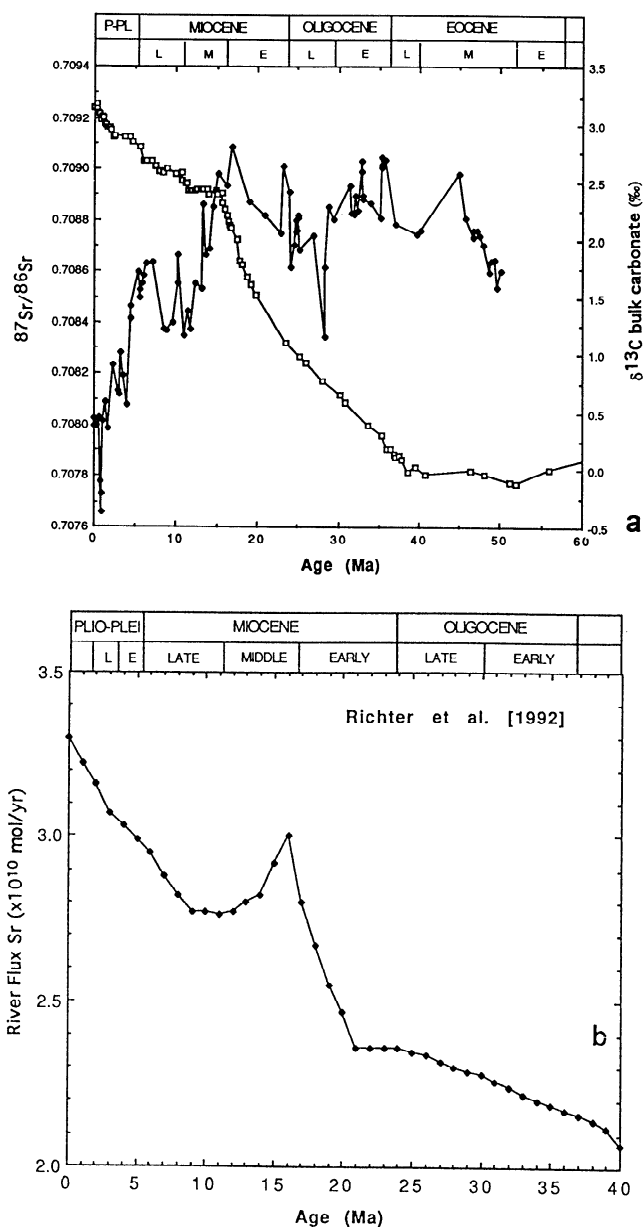


Figure 1. (a) Open squares indicate the strontium isotopic record of global ocean for the last 60 m.y. (Data is from D. DePaolo, private communication, 1992; see also *Richter et al.*, [1992]); solid diamonds indicate the $\delta^{13}\text{C}$ of bulk carbonate from *Shackleton* [1987]. For a discussion of differences between foraminiferal and bulk carbonate $\delta^{13}\text{C}$ records, see *Broecker and Woodruff* [1992]. (b) River strontium flux estimated by *Richter et al.* [1992] for the last 40 m.y. This is the flux needed to reproduce the strontium isotopic record when the hydrothermal strontium flux changes in proportion to seafloor generation rates and the additional river strontium flux is assumed to have a value of 0.713. This study implies an approximately 60% increase in river flux over the last 40 Ma.

of hypotheses have been proposed to explain the deposition and character of the Monterey formation [e.g., *Compton et al.*, 1990; *Vincent and Berger*, 1985; *Vincent et al.*, 1985]. Here it is proposed that deposition of the

lower calcareous organic-rich unit of the Monterey was the result of increased nutrient delivery to the ocean from the Himalayan episode of exhumation and weathering described above. Rather than an increase in trade wind strength and coastal upwelling intensity [Vincent and Berger, 1985] or a rise in sea level [Compton *et al.*, 1990], nutrient loading of upwelling zones may have been responsible for enhanced productivity and OM deposition. DePaolo and Finger [1991] first recognized that a global increase in weathering preceded organic carbon deposition and global cooling and that they may have been related events.

The Organic Carbon Subcycle

The record of marine bulk carbonate $\delta^{13}\text{C}$ can be used to examine the idea that increased river nutrient fluxes led to the deposition of the organic-rich units of the Monterey formation in the early to middle Miocene. Because OM is highly enriched in the light isotope of carbon during photosynthesis, any increase in the fraction of organic carbon removed from the surface carbon reservoir (atmosphere, ocean, biosphere, or soils) will make the remaining inorganic carbon relatively more enriched in the heavy isotope. The isotopic composition of the ocean reservoir is recorded by carbonates with little isotopic fractionation. As can be seen in Figure 1a, the $\delta^{13}\text{C}$ of the carbonate output reservoir generally decreased over the last 17 Ma, suggesting that the fraction of organic carbon versus carbonate being buried has also decreased (or that the fraction of OM weathered has increased) [Shackleton, 1987]. The positive shift in $\delta^{13}\text{C}$ observed between about 20 and 17 Ma in Figure 1a could have been caused by a relatively greater fraction of OM being buried at this time (or a relatively smaller fraction of OM weathered) and, hence, a drawdown in atmospheric CO_2 . After that time, the large negative shift in $\delta^{13}\text{C}$ of bulk carbonate indicates that the organic carbon subcycle generally acted as a source of CO_2 to the ocean-atmosphere system.

In Figure 2a, the fraction of C leaving via organic burial over the last 40 Ma (right axis) is calculated using the following mass balance equation (which sets the flux of C into the ocean-atmosphere equal to the C fluxes out of system in carbonate and organic matter):

$$[\text{Flux}_{\text{river}}] [\delta^{13}\text{C}_{\text{river}}] =$$

$$[\text{Flux}_{\text{carb}}] [\delta^{13}\text{C}_{\text{carb}}] + [\text{Flux}_{\text{org}}] [\delta^{13}\text{C}_{\text{carb}} - 25]$$

The mean river $\delta^{13}\text{C}$ value is assumed to be constant at -3.8‰ [Delaney and Boyle, 1988], while the $\delta^{13}\text{C}$ of organic matter is assumed to be fractionated by -25‰ from carbonate [Shackleton, 1987]. Allowing the photosynthetic fractionation to decrease by a few permil toward the present [Popp *et al.*, 1989] does not significantly affect the results discussed below. The $\delta^{13}\text{C}_{\text{carb}}$ is derived from the Shackleton [1987] data, shown in Figure 1a, interpolated at 1-m.y. increments. The calculation is initiated at 40 Ma when the carbonate burial flux, $\text{Flux}_{\text{carb}}$, is assumed to be 12.5×10^{12} mol/yr (150×10^{18} g C/Ma) at 2.1‰ and the organic carbon flux (24% of total

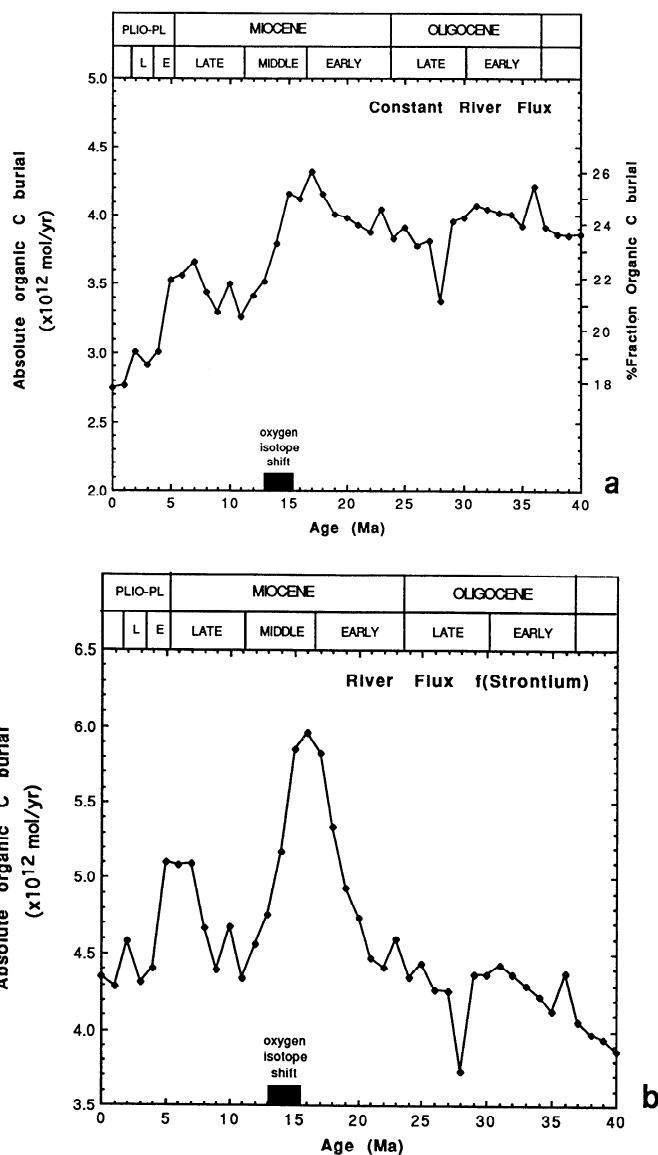


Figure 2. (a) Absolute organic carbon burial for the late Cenozoic calculated using mass balance constraints described in text and assuming a constant river carbon flux. The interval over which the permanent Antarctic ice sheet formed is indicated by the solid bar on age axis. The percent fraction of carbon buried in (or weathered from) an organic reservoir versus a carbonate reservoir is indicated on the right. (b) Absolute organic carbon burial calculated using mass balance constraints and assuming river carbon fluxes are proportional to the change in river strontium fluxes estimated by Richter *et al.* [1992] and shown in Figure 1b. The percent fraction of C leaving as organic matter versus carbonate at any one time does not change from Figure 2a. Note that both curves have the same initial organic matter burial flux.

C flux), Flux_{org} , is assumed to be 3.86×10^{12} mol/yr (46.3×10^{18} g C/Ma) at -22.9‰ . This implies a Cenozoic river flux of carbon of 16.36×10^{12} mol/yr (or 196×10^{18} g C/Ma) which is about 30% higher than the long-term steady state carbon flux used by Shackleton [1987].

Using the above data and assumptions, the percent fraction of C buried as organic matter decreases from approximately 24% to 18% over the last 40 Ma (Figure 2a, right axis). Note that the calculation of the fraction of carbon buried in organic versus inorganic reservoirs is insensitive to changes in the river input flux of carbon, $\text{Flux}_{\text{river}}$. However, to calculate absolute organic carbon burial rates as we wish to do here, one must make an assumption about how river fluxes have changed. *Shackleton* [1987] (and *Vincent and Berger* [1985]) assumed that the flux of C weathered from rock (or released by mantle degassing) was constant. Calculated net organic carbon burial rates shown in Figure 2a (left axis) assume a constant river carbon flux. This curve is essentially identical to that in *Shackleton* [1987]. Note that absolute organic carbon burial rates were not particularly high during the late early to early middle Miocene interval of Monterey organic-rich deposition and, in fact, fell through much of this interval, as dictated by the bulk $\delta^{13}\text{C}_{\text{carb}}$ data.

What happens if river fluxes are allowed to vary? In Figure 2b, the 40 Ma river carbon flux was again assumed to be 16.4×10^{12} mol C/yr, but then it was allowed to increase in proportion to the change in river Sr flux calculated by *Richter et al.* [1992] (Figure 1b). In other words, silicate weathering, carbonate weathering, and OM weathering all increased in proportion to Figure 1b. The carbonate flux value obtained at the end of the calculation (0 Ma) is the modern carbonate burial flux estimate of *Wilkinson and Algeo* [1989], 20×10^{12} mol C/yr. At each 1-m.y. time step, the organic carbon burial flux was determined by mass balance using the above steady state equation. Using the strontium curve as a proxy for river carbon flux, a pronounced increase in absolute organic carbon burial rates is predicted for the late early Miocene to early middle Miocene interval when most of the organic-rich units of the Monterey formation were deposited (Figure 2b). Organic carbon burial rates between 17 and 15 Ma are about 40% higher than "normal" late Cenozoic levels. Likewise, the interval of elevated organic carbon burial between 8 and 5 Ma may have been stimulated by enhanced chemical weathering and river nutrient fluxes associated with the intensification of the Asian monsoon around 8 Ma [*Quade et al.*, 1989; *Prell and Kutzbach*, 1992].

Discussion

This modification of the Monterey hypothesis, whereby tectonically driven exhumation in the Himalayas increases river nutrient fluxes, also accounts for the observed increase in carbonate deposition at this time [*Woodruff and Savin*, 1991], an observation that was at odds with the traditional hypothesis of *Vincent and Berger* [1985]. In addition, the time of most rapid CO_2 drawdown was between 20 and 16 Ma and was driven primarily by increased continental weathering of silicates rather than by an increase in the fraction of organic matter buried versus carbonate. The timing of the CO_2 drawdown predicted here is in almost perfect agreement with paleo- pCO_2 estimates based on OM $\delta^{13}\text{C}$ which show a rapid shift in

atmospheric CO_2 levels, from ~5 times preindustrial levels at 20 Ma to ~1.5 times preindustrial values by 15 Ma [*Arthur et al.*, 1991]. This is also the interval over which cooling and a major phase of ice growth in Antarctica takes place [*Leckie and Webb*, 1983, 1985; *Miller et al.*, 1991; *Oslick et al.*, this issue], culminating in the establishment of a permanent continental ice sheet, as indicated by a positive shift in oxygen isotopes, between 15 and 13 Ma [*Woodruff and Savin*, 1991; *Woodruff et al.*, 1981].

How does the organic carbon subcycle act as a negative feedback to climate change? Clearly, between about 18 and 16 Ma the increased fraction of organic carbon buried relative to carbonate (and relative to OM weathered) exacerbates the problem of CO_2 drawdown driven by tectonically related weathering. However, at 16 Ma a profound change in the behavior of the organic subcycle took place: after this point, and for almost all the rest of the Cenozoic, the percent fraction of organic carbon burial decreased. This implies that although absolute organic carbon burial rates had never been higher, organic matter weathering exceeded organic matter burial, providing a source of CO_2 to the atmosphere to offset removal by silicate weathering. This interpretation is supported by the positive shift in the oceanic osmium isotopic record observed at this time, radiogenic osmium being concentrated in organic-rich sediments [*Ravizza*, 1993].

The drop in sea level associated with the rapid expansion of the Antarctic ice sheet after 16 Ma may have played a role in tipping the organic carbon/carbonate balance. Exposure and erosion of previously deposited organic-rich Monterey sediments on continental shelves could have resulted in the release of CO_2 back into the atmosphere as well as the formation of widespread phosphorite deposits of the middle Miocene with the liberated, nonvolatile, and reworked phosphorus [*Compton et al.*, 1990]. Alternatively, a colder ocean with correspondingly higher oxygen concentrations could have decreased OM preservation, or increased aridity on land could have decreased areal coverage of swamps and marshlands. In addition, the cooler middle Miocene climate could also have suppressed chemical weathering rates in the Himalayas (note drop in river Sr flux in middle Miocene) and/or the high topography created 5 m.y. or so earlier could have been significantly worn down causing silicate weathering rates to decline.

The issue of causality (the chicken-egg dilemma) has been a topic of much discourse (e.g., *Molnar and England* [1990]). The internal consistency of the tectonic, carbon isotopic, strontium isotopic, and sedimentary histories for the early and middle Miocene argues strongly for the causal relationships described above. In particular, the interpretation of the strontium isotopic signal as reflecting (in part) increases in weathering fluxes is supported by the strong correlation between the time of greatest OM deposition predicted by the bulk $\delta^{13}\text{C}$ data (when river C fluxes are assumed proportional to the Sr curve) and the observed distribution of carbonate and organic-rich deposits in the Neogene. Further, the temporal correspondence of the rapid $\delta^{87}\text{Sr}$ increase to the interval

of major movement on the fault systems of the Himalayas suggests that they could be related. If one assumes that 6 km of rock were exhumed and eroded between the Main Central thrust system and the South Tibetan detachment system (an area approximately 1600 km long and 100 km wide) in the early Miocene and one further assumes that approximately 25% of this rock was chemically weathered, had a strontium concentration of 350 ppm and an isotopic ratio of 0.769, this would provide more than enough strontium to account for the seawater evolution curve between 20 and 15 Ma (see also *Richter et al.* [1992]). The above scenario differs from the Monterey hypothesis of Vincent and Berger in that CO₂ drawdown is primarily via silicate weathering rather than organic carbon burial and that organic carbon burial is driven by increased delivery of nutrients to the ocean rather than by stronger upwelling.

By contrast, other climate transitions in the Cenozoic may have different causes. For instance, in the early late Eocene, the ca. 39-40 Ma inflection in the $\delta^{87}\text{Sr}$ record lags the $\delta^{18}\text{O}$ shift as predicted by a causal relationship which has ice sheets increasing chemical weathering rates [*Miller et al.*, 1991; *Armstrong*, 1971]; there is no corresponding event known from the Himalayas or any other mountain range. On the other hand, the shift to more positive $\delta^{87}\text{Sr}$ in the late Miocene (~9-7 Ma) is not associated with any obvious ice growth event, although a global shift in C3 to C4 type plants (as well as Figure 2b) suggests a drop in CO₂ levels occurred at this time [*Cerling et al.*, 1993]. These changes may be related to an increase in the elevation of the Tibetan Plateau and an associated increase in monsoon strength [*Prell and Kutzbach*, 1992; *Quade et al.*, 1989].

The absolute change in river fluxes during the Cenozoic is highly uncertain, and ultimately, this estimate needs to be improved if we are to develop a quantitative understanding of the long-term evolution of CO₂ in the atmosphere. In this model (taking Figure 1 at face value and using the flux values described above) approximately $\sim 5000 \times 10^{15}$ moles C were removed from the ocean-atmosphere reservoir between 13.5 and 17.5 Ma. Given the approximately 3000×10^{15} moles of carbon in the preindustrial ocean and atmosphere, this suggests that either early Miocene atmospheric CO₂ levels were significantly higher than at present [*Arthur et al.*, 1991], the amount of $\delta^{87}\text{Sr}$ change due to increases in weathering was overestimated by *Richter et al.* [1992], or volcanic CO₂ fluxes were higher in the middle Miocene [*Hodell and Woodruff*, this issue]. Detailed measurements of the $\delta^{13}\text{C}$ of organic carbon, the $\delta^{13}\text{C}$ of mean ocean carbonate, atmospheric CO₂, ocean alkalinity by proxy [*Rau et al.*, 1991; *Freeman and Hayes*, 1992; *Spivak et al.*, 1993], and the development of other chemical weathering proxies should provide much-needed constraints on quantitative modeling of the global carbon cycle.

Acknowledgments. I thank those who have discussed this topic with me or read earlier versions of this paper, especially E. Sundquist, P. Molnar, J. Edmond, E. Boyle, D. Hodell, N. Shackleton, J. Compton, B. Garrison, and K. Miller. I would also like to thank the MG+G program at NSF (OCE92-57191) and the

Petroleum Research Fund of the American Chemical Society for financial support.

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(Received October 6, 1993; revised January 27, 1994; accepted January 27, 1994)