CALCULATION OF THE PHANEROZOIC STRONTIUM ISOTOPE RECORD OF THE OCEANS FROM A CARBON CYCLE MODEL

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ABSTRACT. Results of the long term carbon cycle model (Berner, 1991) have been used to calculate the values of ⁸⁷Sr/⁸⁶Sr for seawater over Phanerozoic time. Rates of weathering of silicates and carbonates and the rate of sedimentary burial of carbonates over time are derived directly from the model. The ⁸⁷Sr/⁸⁶Sr ratio of carbonates undergoing weathering and marine burial are fixed by carbonate recycling also according to the model. Exchange of Sr isotopes via basalt-seawater reaction is assumed to vary directly over time with the rate of seafloor spreading, values of which are derived from the sealevel inversion model calculations of Gaffin (1987).

Use of a constant 87Sr/86Sr for silicates undergoing weathering results in a calculated curve which is in bad agreement with measured Phanerozoic oceanic 87Sr/86Sr values for the period of 300 Ma to present, and this disagreement cannot be removed by varying critical parameters of the carbon model such as rates of global degassing or weathering as affected by orogenic uplift. By contrast, use of an expression relating values of the mean isotopic composition of silicates undergoing weathering to changes in eustatic sealevel results in crude first order agreement with the overall trend of the measurements (the silicate Sr isotopic ratio varies from .709 to .716). The expression reflects the reasonable assumption that the production of relatively nonradiogenic, mantlederived silicates (volcanics, volcaniclastic sediments, et cetera) and their subaerial and submarine weathering is greater when sealevel is higher due to increases in seafloor spreading rate. Because sealevel is also affected by continental breakup and collision, the use of sealevel can also serve as a proxy for the addition of radiogenic Sr accompanying the uplift of old rocks during continental collision.

It is likely that trends in the Sr isotopic composition of the oceans over time have been primarily due to changes in the isotopic composition of materials undergoing weathering and not to large changes in

weathering fluxes and/or hydrothermal fluxes.

INTRODUCTION

The strontium isotopic composition, in terms of the ⁸⁷Sr/⁸⁶Sr ratio, for the oceans over Phanerozoic time has been rather well established from the analysis of limestones (Burke and others, 1982). Interpretation of this ratio has led to a number of modeling attempts to calculate rates of weathering on the continents and rates of basalt-seawater reaction in the oceans at present and over geologic time (Brass, 1976; Spooner, 1976; Albarede and others, 1981; Holland, 1984; Wadleigh, Veizer, and Brooks, 1985; Chaudhari and Clauer, 1986; Hess, Bender, and Schilling, 1986; Goldstein and Jacobsen, 1987; Palmer and Edmond, 1989; Kump, 1989; Raymo, 1991). Unfortunately there is no unique solution because there

are not enough equations to allow solution for each variable. This is especially true for the problem of changes in the mean ⁸⁷Sr/⁸⁶Sr ratio of the source rocks undergoing weathering versus changes in the rates of weathering and/or basalt-seawater reaction. A sensitivity analysis to each of the possible variables in the mass balance expressions for total strontium and for radiogenic ⁸⁷Sr (Kump, 1989) illustrates very well the non-uniqueness of any one solution to the problem.

One can constrain Sr isotope modeling by relating it to results of the modeling of other elements which has not been done in the earlier studies cited above. Here we do this by using the results of carbon cycle model calculations for the Phanerozoic (Berner, 1991). One result of the carbon modeling was calculation of the rates of carbonate and silicate weathering and rates of carbonate sediment burial over the past 570 my. Since the Sr/(Ca + Mg) ratios of silicates and carbonates are about the same (Wedepohl, 1978), one can get some idea of rates of supply of Sr by rivers to the ocean over time from the carbon modeling. This is done in the present paper in terms of a simple model which incorporates outputs from the carbon model with changes in the Sr isotopic composition of silicates and carbonates undergoing weathering in order to match the Phanerozoic oceanic 87Sr/86Sr record. Results show that the modeling of both the cycles of carbon and strontium can lead to compatible results if one assumes that the mean Sr isotopic composition of rocks undergoing chemical weathering varies with time due to global tectonic factors.

ST ISOTOPE MODEL

The Sr isotope modeling used here assumes that the ⁸⁷Sr/⁸⁶Sr composition of the oceans can be treated as a succession of steady states for Sr. For rapid changes this is inappropriate because of the approx 3 my residence time of Sr in the oceans. However, for emphasis on the first order pattern of gradual changes over Phanerozoic time, this approach is sufficient. Accordingly the appropriate mass balance expression for ⁸⁷Sr is:

$$R_{c}r_{c}F_{wc} + R_{s}r_{s}F_{ws} + R_{b}r_{b}F_{bo} = R_{o}r_{c}F_{bc} + R_{o}r_{ab}F_{ob}$$
 (1)

where:

 F_{wc} = weathering flux of Ca + Mg from carbonates

 F_{ws} = weathering flux of Ca + Mg from silicates

 F_{bc} = sedimentary burial flux of Ca + Mg carbonates in the oceans

 F_{bo} = rate of transfer of Ca + Mg from basalt to the oceans due to reactions along mid-ocean rises

F_{ob} = rate of transfer of Ca + Mg from seawater to basalt along the mid-ocean rises

 $R_c = {}^{87}Sr/{}^{86}Sr$ of carbonates undergoing weathering

 $R_s = {}^{87}Sr/{}^{86}Sr$ of silicates undergoing weathering

 $R_o = {}^{87}Sr/{}^{86}Sr$ of the ocean

 $R_b^{\circ} = {}^{87}Sr/{}^{86}Sr$ of mid-ocean basalts

 $r_c = ratio of ^{86}Sr to Ca + Mg in carbonates undergoing weathering or$ ultimate burial (diagenetic changes in Sr are ignored—see Kump, 1989)

r_s = ratio of ⁸⁶Sr to Ca + Mg in silicates undergoing weathering

 r_b = ratio of 86 Sr to Ca + Mg in fresh midocean basalts r_{ab} = ratio of 86 Sr to Ca + Mg in hydrothermally altered midocean basalts

As mentioned above, the Sr/(Ca + Mg) ratio of silicates and carbonates (after diagenesis) are approximately equal. Thus, as a first approximation one can assume that $r_c = r_s = r_b = r_{ab}$. Also, most workers agree that there is no net uptake or release of total strontium via basaltseawater reaction but only isotope exchange (Albarede and others, 1981). Thus, it can be assumed that $r_{ab}F_{ob} = r_bF_{bo}$. If $r_{ab} = r_b$, then $F_{ob} = F_{bo}$. For changes in F_{ob} with time due to changes in seaffoor spreading rate we use the expression:

$$\mathbf{F}_{ob} = \mathbf{f}_{sr}(\mathbf{t})\mathbf{F}_{ob}(0) \tag{2}$$

where:

 $F_{ob}(0)$ = the rate of exchange of Ca + Mg between midocean basalt and seawater at present

 $f_{sr}(t)$ = ratio of rate of seafloor spreading at a given time in the past to that at present (see Berner, Lasaga, and Garrels, 1983)

Substituting eq (2) and the above arguments into eq (1) we can solve for the ⁸⁷Sr/⁸⁶Sr ratio of seawater R₀ as a function of time:

$$R_{o} = [R_{c}F_{wc} + R_{s}F_{ws} + R_{b}f_{sr}(t)F_{ob}(0)]/[F_{bc} + f_{sr}(t)F_{ob}(0)]$$
(3)

Values of f_{er}(t) are derived from the eustatic sealevel inversion calculations of Gaffin (1987) (see also Berner, 1991). The value of F_{ob}(0) is taken as 6.5×10^{18} moles per million years from the hydrothermal flux value of Palmer and Edmond (1989) and the Mg content of seawater. (Use of alternate values for this flux—for example, Wolery and Sleep, 1988—have little effect on results). Values of F_{wc} , F_{ws} , and F_{bc} as a function of time are taken directly from the computer output of the carbon model (Berner, 1991). The value of R_b is assumed to be .703 in agreement with earlier studies. The value of R was allowed to vary with time as carbonates undergo burial and recycling via weathering and metamorphic breakdown. To accomplish this an expression analogous to that used for C isotopes was employed:

$$dR_c/dt = (R_o - R_c)F_{bc}/C \tag{4}$$

where:

C = global mass of carbon present as carbonates at any one time.

Elucidation of a value for R_s is more problematical. It can be held constant over time or allowed to vary as the proportions of radiogenic • and nonradiogenic silicates undergoing weathering vary. Here we will examine both scenarios and show that letting R_s vary over time is necessary to reconcile strontium isotope modeling with the results of the carbon model. We will let R_s vary with paleo-sealevel assuming that over the long term sealevel is affected by continental collision and breakup and/or by changes in seafloor spreading rate. According to this idea, changes in the mean isotopic composition of silicates undergoing weathering should accompany global tectonic changes which are reflected by changes in sealevel. A more detailed discussion of the topic of silicate weathering can be found in the next section.

The model was run as an addition to the computer program for the Phanerozoic carbon cycle model (Berner, 1991). Details of calculation can be found in this paper. In summary, the carbon model rests on two mass balance expressions for the input and output of total carbon and 18 C to and from the ocean-atmosphere system. This enables calculation of the weathering fluxes of silicates, carbonates, and organic matter, the deposition fluxes of carbonates and organic matter in sediments, and the volcanic-metamorphic degassing fluxes from the thermal breakdown of buried carbonates and organic matter. The weathering flux for silicates (F_{ws} above) is calculated as the difference between the burial and weathering fluxes for carbonates. This difference represents the transformation of silicates to carbonates as a result of Ca-Mg silicate weathering followed by Ca-Mg carbonate precipitation in the oceans.

In the present study the initial Sr isotopic values at the beginning of the Cambrian were chosen so as to result in the *approximate* Sr isotopic value of the present-day ocean and the *approximate* average Sr isotopic ratio for Phanerozoic carbonates as they exist today. (Attempts to reproduce precisely present day values were felt to be unnecessary considering

large errors in other aspects of the carbon model.)

RESULTS AND DISCUSSION

Plots of eq (3) as a function of time are shown in figures 1 to 5. In figure 1 the standard results for F_{wc} , F_{ws} , and F_{bc} from the carbon model are used along with a constant value of $R_s = .716$. As can be seen there is a crude first-order fit to the overall trend of measurements from 570 to about 300 myBP, but not afterward. (Attempts to fit shorter term fluctuation in $^{87}Sr/^{86}Sr$ are beyond the scope of the carbon model and furthermore require a non-steady state approach that is not used here.)

Attempts to get a better fit between theory and measurements by varying a number of key parameters in the carbon model are shown in figures 2 to 4. Figure 2 shows that holding the rate of seafloor spreading constant, in other words $f_{sr}(t) = 1$, for all time, has little effect on results. In fact letting $f_{sr}(t)$ vary over a broad range also has little effect. This is because in the carbon model faster basalt-seawater reactions due to increases in $f_{sr}(t)$ are counterbalanced by increases in GO_2 degassing. Degassing rate is also assumed to be proportional to $f_{sr}(t)$ and higher degassing results in faster weathering of rocks on the continents. This

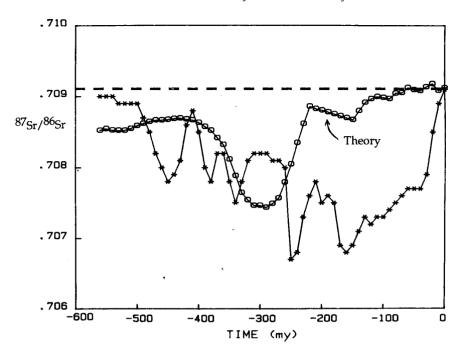


Fig. 1. Plots of ⁸⁷Sr/⁸⁶Sr ratio of the ocean versus time for theoretical calculation compared to the measurements summarized by Burke and others (1982). Theoretical calculation based on the standard formulation of the carbon cycle model of Berner (1991) and the assumption that the ⁸⁷Sr/⁸⁶Sr ratio of silicates undergoing weathering is held constant over time at .716.

comes about because on a million year time scale (see Berner, 1991) one cannot store CO_2 in the ocean-atmosphere system. If more CO_2 is added to the atmosphere/ocean by degassing, this CO_2 must be removed via weathering in order to prevent the buildup of inordinately high CO_2 levels at the Earth's surface. Since the weathering flux is more radiogenic than seawater and that from basalt-seawater reaction less radiogenic, increases in both processes with increase in $f_{\rm sr}(t)$ tend to cancel one another. One can partly get around this problem by calling on the weathering of seafloor basalt to act as a principal control on atmospheric CO_2 . However, this depends on a much better understanding of seafloor weathering and its quantitative significance to atmospheric CO_2 levels which has proven to be controversial (see Staudigel and others, 1989; Berner, 1990; Caldeira, 1991).

Figure 3 shows results of holding all tectonic and biological factors affecting global weathering and degassing constant in the carbon model. This represents the situation of no change in the area, mean elevation, or position of the continents with time, no change in weathering rate as the result of the rise and evolution of vascular land plants, and no change in

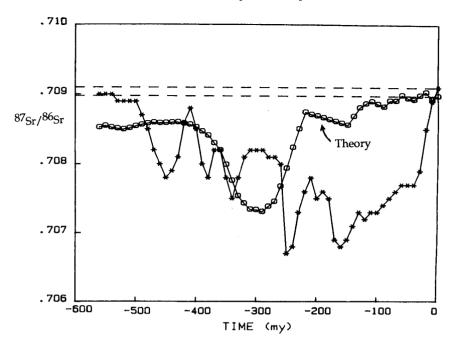


Fig. 2. Same situation as for figure 1 except that the rate of seafloor spreading $f_{sr}(t)$ is held constant and equal to one for all time.

 ${\rm CO_2}$ degassing rate due to changes in spreading rate. In this case the only factor affecting ${\rm F_{wc}}$, ${\rm F_{ws}}$, and ${\rm F_{bc}}$ is the rate of burial of organic matter as governed by the carbon isotopic record of the oceans. As can be seen there is merely an upward shift of the calculated ${\rm ^{87}Sr/^{86}Sr}$ curve with no appreciable change in its shape.

Figure 4 is for the opposite situation. This is for a constant carbon isotopic composition of the oceans over Phanerozoic time but inclusion of all other factors affecting the weathering and burial of carbonates and silicates. Results show that the theoretical curve nowhere fits the general trend of the Sr isotopic data. This indicates that the general shape of the curve in figure 1 for the early to mid-Paleozoic, and its crude match to observations, is due mainly to changes in the burial rate of organic matter (and carbonates) as reflected by changes in oceanic carbon isotope composition. Increased burial of organic carbon, accompanying increased δ^{13} C of the shallow ocean in the mid-Paleozoic, entailed reduced burial of carbonates to maintain isotope mass balance. This decrease in carbonate burial, according to the carbon model (Berner, 1991), necessitated a decrease in Ca-Mg silicate weathering which resulted in a mid-Paleozoic drop in the river input of radiogenic Sr to the oceans.

Raymo (1991) has recently suggested that changes in weathering rate due to uplift of rocks during orogenies should affect both atmo-

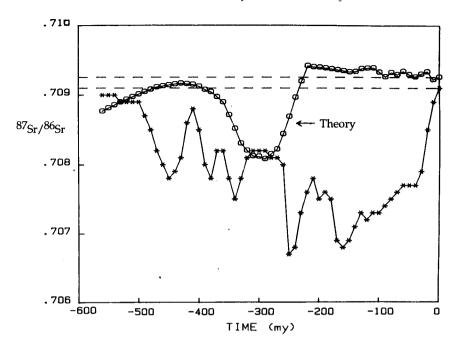


Fig. 3. Same situation as for figure 1 except that all factors affecting weathering and global CO_2 degassing in the carbon cycle model are held constant over time. This means that the area, mean elevation, and position of the continents, the rate of seafloor spreading, and the distribution of carbonates between platforms and the deep sea are all held constant and that there are no effects on weathering due to the rise and evolution of vascular land plants.

spheric CO₂ and the Sr isotopic composition of seawater. Increases in oceanic ⁸⁷Sr/⁸⁶Sr, caused by increased continental weathering, should be accompanied by drops in atmospheric CO₂. However, use of different and strongly varying values of $f_p(t)$, the term expressing the role of mean continental elevation on weathering rate (Berner, 1991), results in predicted plots of ⁸⁷Sr/⁸⁶Sr which are essentially the same as that shown in figure 1. This comes about because the Raymo reasoning does not take into account the necessity of balancing, on the scale of millions of years, the weathering uptake of CO₂ with the input of CO₂ to the atmosphere by metamorphic/volcanic degassing. Certainly, at constant degassing an increase in weathering rate due to uplift is accompanied by a decrease in atmospheric CO₂, but if there is no corresponding increase in the rate of CO₂ input to the atmosphere, there can be no *sustained* increase in its rate of removal by silicate weathering. Thus, over long time scales uplift alone cannot bring about an increased weathering flux of radiogenic strontium to the sea; it can only result in a drop in atmospheric CO2. Continued increased weathering unaccompanied by equivalent degassing will ex-

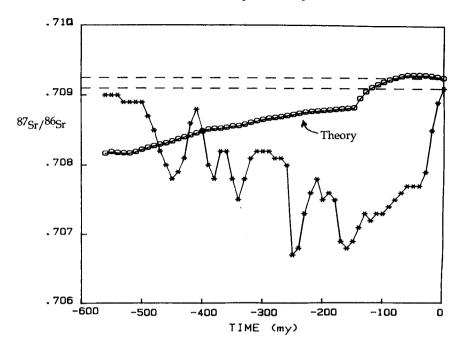


Fig. 4. Same situation as in figure 1 except that the carbon isotopic composition of the oceans in the carbon cycle model is assumed constant at $\delta^{19}C = 1.0$ permil for all time. All other factors are as in the standard formulation (Berner, 1991). The large difference between the theoretical curve of figure 4 and those of figures 1 to 3 can be ascribed to the importance of changes in oceanic $\delta^{19}C$ as they affect the rates of weathering of carbonates and silicates and the sedimentary burial of carbonates.

haust the available CO_2 in the oceans and atmosphere in less than a million years (Sundquist, 1991). As stated above, to maintain carbon balance at the Earth's surface removal of CO_2 by weathering must be balanced on a million year time scale by inputs due to degassing.

At this point one can take two approaches. The first one is to hold R_s essentially constant over time which results in a fundamental inconsistency between previous carbon cycle modeling and the marine Sr isotopic record as shown above and further emphasized by François and Walker (1991). Here, following the lead of Brass (1976), we instead let the value of R_s vary with time. Since the amount of weathering of less radiogenic silicates (such as basalts and volcaniclastic sediments) relative to more radiogenic silicates (such as old cratons subject to uplift and/or glaciation) should have varied over the Phanerozoic, it seems to us to be quite reasonable to let R_s vary with time.

One could simply fit the Sr isotopic record by appropriate variation of R_s , but to retain more theoretical power we have adopted another approach. We assume that the isotopic composition of Sr delivered by

rivers to the sea from continental weathering is a function, albeit indirectly, of sealevel. We assume that changes in sealevel on the long term, are controlled primarily by two tectonic processes: changes in the rate of seafloor spreading along oceanic rises and the collision and breakup of continents. The reasoning goes as follows. First, as spreading rate increases, sealevel rises and global volcanism increases. As a result the abundance of low R_s (mantle-derived) volcanic material available for subaerial weathering, especially along active margins, also increases. This includes both true volcanics and volcaniclastic sediments derived from them. Increases in volcanics available for weathering should cause the global mean value for R_s to decrease. Furthermore, increases in spreading rate should produce greater submarine weathering of basalts (Staudigel and others, 1989) which would also introduce more non-radiogenic Sr to the ocean. Secondly, as continents collide sealevel goes down (Heller and Angevine, 1985; Nance, Worsley, and Moody, 1988), and old radiogenic rocks are uplifted, glaciated, and become available for weathering. In this case the increased weathering of the old rocks should lead to an increase in R. (Brass, 1976; Edmond and Palmer, 1990; Hodell, Mead, and Mueller, 1990). Finally, a rise in sealevel due to continental breakup should result in increased basaltic volcanism along the zones of separation and, upon weathering of this material, lower values of R.

It should be noted that changes in the relative proportions of radiogenic and non-radiogenic silicates undergoing weathering would occur mainly in and near uplifted orogenic mountain belts and plateaus where major silicate weathering takes place (at present in the Andes, Himalayas, and Tibetan Plateau—see Berner and Berner, 1987). Thus, one should not use past changes in relative land areas underlain simply by "young" and "old" rocks to test the idea of changes in R_s. It is the mountainous, orogenic areas where most silicate weathering takes place and use of relative proportions of areas of low-lying and relatively unreactive shields to calculate Sr istopic source compositions is misleading. On the other hand, chemical weathering of carbonates is relatively independent of elevation (Holland, 1984), and the relative exposure areas of carbonates, relative to (uplifted) silicates (Bluth and Kump, 1991), is a factor not considered in the present modeling but which should be in future work.

In order to express these processes and to obtain the best fit possible using the standard formulation of the carbon model, we have employed the following equation:

$$R_s = .716 + 0.01[1 - f_s(t)] \tag{5}$$

where $f_s(t)$ is a dimensionless variable representing (tectonically-controlled) sealevel as a function of time relative to that of today. Rather than using sealevel directly, we allow for time lags in sealevel change resulting from global tectonic events. This is the approach of Gaffin (1987), and as a first order approach to the problem, we simply use Gaffin's inversion to.

apply to both collision/breakup and spreading rate effects. In other words we assume that $f_s(t) = f_{ss}(t)$.

Eq (5) results in values for R_s ranging from .716 to .709 over Phanerozoic time. To show that these values are reasonable the following calculation for modern weathering is made. We assume that $r_c = r_s$ and that 66 percent of Ca + Mg in rivers worldwide is derived from carbonate weathering and 34 percent from silicate weathering (Berner, Lasaga, and Garrels, 1983). We also assume that the value of R for present day carbonate weathering is .708, representing a mean for the past 100 my. Combining these estimates with two recent estimates of the mean value of ⁸⁷Sr/⁸⁶Sr for global average river water .7119 (Palmer and Edmond, 1990) and .7101 (Goldstein and Jacobsen, 1987), we obtain values of R for today of .719 and .714 respectively. From eq (5) the value of R_s for today ($f_{sr}(t) = 1$) is .716 which falls in the middle. However, the river value of Palmer and Edmond is probably more accurate, as it is based on more data, and if so, our equation falls somewhat short of predicting the present day value for R. However, we feel that overall eq (5) is a reasonable first order representation of the effect of tectonism and volcanism (as mirrored by sealevel changes) on R, over Phanerozoic time.

A plot of the curve of ⁸⁷Sr/⁸⁶Sr, theoretically predicted using eq (5) and the standard formulation of the carbon model, is shown in figure 5. As one can see there is a crude fit to the general overall U-shaped trend of the measured data, much better than that resulting when one assumes a fixed value for R_s (figs. 1–4). Major discrepancies do occur during the Mesozoic, between 240 and 100 myBp, which can be explained in terms of additional factors.

Discrepancies such as those found for the Mesozoic most likely reflect the fact that orogenic belts are different, both in their makeup and in their development through time, and that changes in the proportions of different types can be seen in terms of changes in the global average isotopic composition of silicates undergoing weathering, R_s. In our model we have considered that orogenic belts dominate silicate weathering and that they form during subduction or during continental collisions. Those belts that form from subduction processes are related to increased spreading rates, higher sealevel, and less radiogenic Sr, whereas those that form by continental collision are related to lower sealevel and more radiogenic Sr. Other scenarios are clearly possible. For instance, collision between a large oceanic arc and a young continental margin could well produce lower sealevel and unradiogenic Sr.

A further oversimplification of our model is that we have not considered the fact that as an orogenic belt ages the Sr isotopic ratio will vary, both as radiogenic Sr is produced and as different rock types become exposed to weathering. If we consider, for example, the Mesozoic orogenic belt of California, one might expect that during initial uplift old preexisting continental rocks dominated silicate weathering. However, the rocks that are now found as wall rocks originally had ⁸⁷Sr/⁸⁶Sr ratios between .704 and .738 (Kistler and Peterman, 1973) and ranged in age

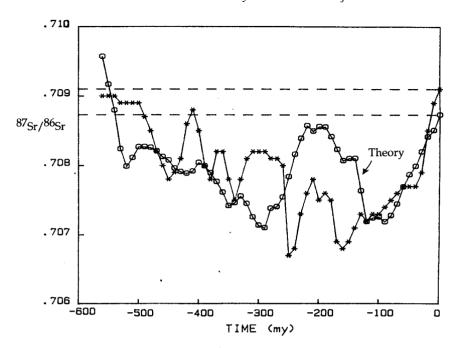


Fig. 5. Plots of ${}^{87}\mathrm{Sr}/{}^{86}\mathrm{Sr}$ ratio of the oceans versus time for theoretical calculation compared to the measurements summarized by Burke and others (1982). Theoretical calculation is based on the standard formulation of the carbon cycle (Berner, 1991) using eq (5) for the value of the ${}^{87}\mathrm{Sr}/{}^{86}\mathrm{Sn}$ ratio of silicates undergoing weathering. This expresses the variability over time of the global mean Sr isotopic composition of silicates undergoing weathering.

from late Paleozoic to Precambrian. As the volcanic edifice grew the Sr isotopic composition was dominated by rocks with isotopic ratios between .703 and .708 (Kistler and Peterman, 1973). As the volcanic edifice was removed by erosion the isotopic composition of silicates probably became more radiogenic as more old rocks were intersected in the roof pendants. Eventually, as the Mesozoic batholiths were intersected, the isotopic ratios became dominated by rocks that had 87Sr/86Sr also in the range of .703 to .708. (Note here the relatively nonradiogenic character of both volcanics and granite.) In the future one expects that the Sr being weathered in the California Mesozoic orogenic belt from silicates will become more radiogenic for two reasons. First, the 87Sr/86Sr ratio in the granites is increasing by about .001/100 my due to radioactive decay (Kistler and Peterman, 1973), and second, as the belt is dissected further a larger amount of radiogenic wall rocks in the form of high grade metamorphic rocks will begin to weather. This all shows how complex the evolution of orogenic belts can be as their weathering affects the Sr isotopic composition of seawater.

Finally, there are other factors that could explain disgreements between our predicted and the measured Sr isotopic values. For one, the assumption that r (the ratio of Sr to Ca + Mg) is constant for all rocks and does not vary with time is crude and needs further study. Also, if the reaction between basalt and seawater at oceanic rises results in some net input or output of Sr from the oceans this could also affect results.

CONCLUSIONS

A crude first order fit to the general trend of oceanic ⁸⁷Sr/⁸⁶Sr over Phanerozoic time can be made by combining results from the carbon model of Berner (1991) for rates of weathering of silicates and carbonates and rates of burial of carbonates with the assumption that the global mean value of ⁸⁷Sr/⁸⁶Sr for Ca-Mg silicates undergoing weathering varies with time. To formulate the latter the relative availability to weathering of relatively non-radiogenic volcanics and old radiogenic silicates in orogenic belts is set proportional to changes in sealevel. Improvements in our approach will come with a greater understanding of the relation between the isotopic composition of Sr delivered by rivers to the oceans and global tectonics. At any rate, we feel that previous carbon cycle modeling is not necessarily incompatible with the marine Sr isotopic record.

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