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A normalised seawater strontium isotope curve and the Neoproterozoic-Cambrian chemical weathering event

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Abstract

The strontium isotope composition of seawater has varied over geological time in response to changes in the rates of continental weathering relative to ocean crust alteration. However, the potential of the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ Sr curve to trace globally integrated chemical weathering rates has not been fully realised because ocean $^{87}\text{Sr}/^{86}\text{Sr}$ is also strongly influenced by the isotopic evolution of Sr sources to the ocean. A first attempt is made here to normalise the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curve to plausible trends in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the three major Sr sources: carbonate dissolution, silicate weathering and submarine hydrothermal exchange. The normalised curve highlights the Neoproterozoic-Cambrian interval as a period of exceptionally high chemical weathering rates, which can be linked to increased nutrient availability, bioproductivity and oxygenation of Earth's surface environment. Use of normalised seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curves will, it is hoped, help to improve future geochemical models of Earth System dynamics.

1 Introduction

Strontium in modern seawater ($^{87}\text{Sr}/^{86}\text{Sr} \sim 0.709$) derives from two major sources: the submarine, chemical alteration of ocean crust ($^{87}\text{Sr}/^{86}\text{Sr} \sim 0.703$: Hofmann, 1997) and the subaerial, chemical weathering of the continental crust and its sedimentary cover ($^{87}\text{Sr}/^{86}\text{Sr} \sim 0.712$: Palmer and Edmond, 1989; Peucker-Ehrenbrink and Miller, 2006). The $^{87}\text{Sr}/^{86}\text{Sr}$ signatures of these two isotopic end members indicate that about twice as much ocean Sr derives from river runoff (RR) than from hydrothermal exchange (MI). Because seafloor spreading rates, which determine ocean crust production, are not considered to change erratically over geological time (Rowley, 2002), attempts have been made to use the marine carbonate-based seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curve to trace changes in globally integrated chemical weathering rates (Francois and Walker, 1992; Kennedy et al., 2006). This approach is complicated, however, by the fact that seawater

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$^{87}\text{Sr}/^{86}\text{Sr}$ is affected not only by changes in the rates of continental weathering relative to sea-floor spreading but also by variations in the isotopic composition of river runoff and the upper mantle. Uncertainties in the $^{87}\text{Sr}/^{86}\text{Sr}$ of past river runoff, in particular, undermine the potential of the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curve to be an unambiguous tracer of chemical weathering (Veizer and Mackenzie, 2003).

The $^{87}\text{Sr}/^{86}\text{Sr}$ signature of RR is a function of the ratio between carbonate and silicate weathering rates with respect to Sr. Although it is widely held that carbonate weathering is the dominant source of Sr in river runoff (75%: Brass, 1976; 67%: Berner and Rye, 1992; 78%: Veizer and Mackenzie, 2003), variations in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the carbonate component in river runoff have generally not been considered in interpretations of first order seawater $^{87}\text{Sr}/^{86}\text{Sr}$ evolution. Here I propose plausible, but not incontrovertible evolutionary trends for the three major Sr sources to the ocean: 1) submarine, elemental exchange with ocean crust (MI); 2) subaerial, chemical weathering of silicate minerals (RRS); and 3) subaerial dissolution of marine carbonate (and marine sulphate) rocks (RRC), in order to illustrate how the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curve can be normalised against the isotopic evolution of Sr sources to the ocean. The use of more sophisticated, normalised seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curves will it is hoped lead to improved biogeochemical models of the exogenic Earth system.

2 Evolving $^{87}\text{Sr}/^{86}\text{Sr}$ signatures of Sr sources to the ocean

1) Exchange between seawater and ocean crust (MI in Fig. 1) occurs generally at mid-ocean ridges and ridge flanks and supplies Sr to the ocean that inherits the isotopic composition of its magmatic precursor, the depleted mantle (Spooner, 1976), which is ~ 0.703 (Hofmann, 1997). Modern MI represents the accumulation of radiogenic ^{87}Sr in the depleted mantle, which deviated early in Earth history from the bulk earth $^{87}\text{Sr}/^{86}\text{Sr}$ trend as a result of the preferential incorporation of Rb over Sr into the crust. This process led to a lessening of the rate of $^{87}\text{Sr}/^{86}\text{Sr}$ increase in MI, either since the onset of modern tectonics close to the Archaean-Proterozoic boundary (O’Nions, 1979) or

according to some estimates already by c. 3.8 Ga (McCulloch, 1994).

2) Silicate weathering (RRS in Fig. 1) supplies relatively radiogenic Sr to the oceans by virtue of the high Rb/Sr ratios of most rock-forming silicate minerals and the high mean age of the Upper Continental Crust (UCC). However, it has been noted by several authors that the average isotopic composition of surface rocks exposed to weathering appears to be less radiogenic than would normally be expected simply from the radioactive decay of ^{87}Rb , e.g. ~ 0.716 (Goldstein and Jacobsen, 1988) instead of the expected value of >0.730 (Veizer and Mackenzie, 2003), meaning that radiogenic Sr has been lost from the silicate pile over time (Goldstein, 1988). The mean $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the detrital silicate component in modern rivers is 0.7178 (Bickle, 1994), and this is accepted here to be the best estimate for the isotopic signature of silicate-derived, dissolved Sr in modern rivers (RRS). Tracing the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of RRS into the past is difficult because it depends not only on the isotopic evolution of the upper crust but also on the relative susceptibility to weathering, age and Sr content of the various silicate minerals that make up the exposed parts of the crust.

The Sr isotopic evolution of crustal silicates would normally be expected to deviate from that of the depleted mantle as a mirror image, albeit with a steeper gradient, taking into account the relative size differences of these two Sr reservoirs (O’Nions et al., 1979). However, there is a tendency for Rb and Sr to partition into different mineral phases in the surface environment, which complicates any simplistic trend. Sr is leached preferentially from silicates during chemical weathering and tends to accumulate conservatively in seawater, whereas Rb, although soluble, has a strong affinity to the clay silicate fractions of both soils and fine-grained marine sediments. Although some current thinking (e.g. Kemp et al., 2006) considers that any such surface effects were more than compensated for by the preferential partitioning of Rb into granitic melts during partial melting and fractional crystallisation, there is no consensus on the evolution of crustal $^{87}\text{Sr}/^{86}\text{Sr}$ and it is not the aim of this article to contribute to this complex debate. Accordingly, the RRS trend in Fig. 1a is an idealised curve; however, this should not greatly affect the conclusions of the present article. For example, were

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I to assume unchanging RRS $^{87}\text{Sr}/^{86}\text{Sr}$ through time (Goldstein, 1988), this would only dampen the major features of the normalised curve shown in Fig. 1b but not eliminate them.

3) The dissolution of carbonate rocks (limestones and dolomites) and evaporitic sulphates (gypsum, anhydrite) is considered to be the major source of Sr to the oceans (Brass, 1976) by virtue of the high Sr contents and high solubility of those minerals. The isotopic signature of this largely carbonate source today (~ 0.708) is close to the range of Phanerozoic seawater (~ 0.707 – 0.709) and so changes in the rates and isotopic composition of the carbonate weathering flux (RRC) are widely disregarded in interpretations of ocean $^{87}\text{Sr}/^{86}\text{Sr}$ trends (Veizer and Mackenzie, 2003). However, the $^{87}\text{Sr}/^{86}\text{Sr}$ of RRC is largely dependent on past seawater $^{87}\text{Sr}/^{86}\text{Sr}$ and so will have evolved considerably over time. According to recent estimates (Peucker-Ehrenbrink and Miller, 2006), about half of all sediments undergoing weathering at the Earth's surface are younger than 0.25 Ga, while the rest must be aged between 0.25 and 3.5 Ga with a strong bias towards younger Phanerozoic sedimentary rocks. Assuming that this skewed age distribution has remained the same since carbonate platforms first became abundant around 2.5 Ga, the $^{87}\text{Sr}/^{86}\text{Sr}$ of RRC can be traced back through time (Fig. 1a). Clearly, the changing seawater $^{87}\text{Sr}/^{86}\text{Sr}$ will have exerted considerable control over the isotopic evolution of river runoff independently of the age distribution of rocks undergoing weathering or crustal evolution. Significantly, seawater and predicted RRC $^{87}\text{Sr}/^{86}\text{Sr}$ reached a maximum divergence of ~ 0.004 at around 0.5 Ga (Middle Cambrian–Early Ordovician).

3 River runoff versus mantle input through time

Modern-day river runoff $^{87}\text{Sr}/^{86}\text{Sr}$, at 0.7124 (Palmer and Edmond, 1989; Peucker-Ehrenbrink and Miller, 2006), reflects a mixture of Sr from the weathering of carbonates ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7077$) and silicates ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7178$). Using these estimates, approximately 55% of the Sr in modern rivers derives from the weathering of carbonate

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minerals (cf. 60%: Bickle, 1994). This appears to be a conservative estimate, being considerably lower than the initially proposed 75% ratio (Brass, 1976) and is also lower than the 67% ratio used in published models of the exogenic Sr isotope system (Francois and Walker, 1992; Berner and Rye, 1992). In this regard, it can be argued that the estimated isotopic composition of RRW is only a minimum constraint as marine carbonate and evaporite minerals generally incorporate more radiogenic Sr during diagenetic alteration. The isotopic composition of the modern RRS flux is not well constrained. In particular, the assumption that the Sr isotopic composition of detrital silicates accurately represents that of the dissolved Sr component (Bickle, 1994) may overestimate the $^{87}\text{Sr}/^{86}\text{Sr}$ of RRS due to the generally greater susceptibility to weathering of less radiogenic silicate minerals.

In order to use the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curve as a measure of changes in river runoff (RR) versus mantle Sr input (MI), I have first estimated the $^{87}\text{Sr}/^{86}\text{Sr}$ of RR by assuming that the modern ratio of 45:55 between the silicate and carbonate weathering fluxes has remained the same throughout the past 2.5 Ga. This assumption appears to be somewhat valid as the resultant curve of river runoff $^{87}\text{Sr}/^{86}\text{Sr}$ (Fig. 1a) does not cross even the highest peaks of the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ trend. Indeed, the relative influence of carbonate weathering on the isotopic composition of river runoff is likely to have been even greater in the geological past. This is because carbonate dissolution is not constrained directly by the endogenic (volcanic and metamorphic) CO_2 flux unlike silicate weathering, and was probably favoured in the geological past due to the greater acidity of rainfall caused by higher atmospheric CO_2 (Kasting and Siefert, 2004), while the absence of land plants and eukaryotic soil biota meant that silicate weathering efficiency was likely depressed during the Precambrian relative to today (Lenton and Watson, 2004). The flux of Sr derived from carbonate dissolution seems therefore likely to have dominated over that from silicate weathering for as long as carbonate platforms have been abundant on Earth, i.e. since the early Proterozoic or possibly the late Archean. The relative influence of river runoff versus mantle input (Fig. 1b) can now be estimated by considering seawater $^{87}\text{Sr}/^{86}\text{Sr}$ to be a binary mixture of RR and

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MI.

The resultant normalised $^{87}\text{Sr}/^{86}\text{Sr}$ curve in figure 1b displays some significant deviations from the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curve from which it was derived, and therefore from previous estimates of river runoff versus mantle input. These differences are not particularly sensitive to the age model chosen for carbonate sediments undergoing weathering or to the evolutionary model for the silicate weathering component in rivers.

Most importantly, there is no longer any evidence for a stepwise change in the $^{87}\text{Sr}/^{86}\text{Sr}$ base-level (Fig. 1a) across the Proterozoic-Phanerozoic transition, which had been interpreted previously to indicate an irreversible shift in chemical weathering rates due to the evolution of soil biota on the continents (Kennedy et al., 2006). The shift in the base-level noted by those authors can instead be explained by increases in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the Phanerozoic carbonate weathering and mantle fluxes. Another feature of note is the relative deflation of the Cenozoic $^{87}\text{Sr}/^{86}\text{Sr}$ peak compared with the previously equivalent Cambrian $^{87}\text{Sr}/^{86}\text{Sr}$ peak. The significance of the Neoproterozoic-Cambrian rise in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ can only be fully appreciated when one considers that the $^{87}\text{Sr}/^{86}\text{Sr}$ compositions of both the MI and the RRC fluxes would have been considerably lower during the Neoproterozoic-Cambrian interval.

4 The Neoproterozoic-Cambrian chemical weathering event

The Neoproterozoic-Cambrian rise in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ (normalised or otherwise) can be explained by one or more of four different factors: 1) an increase in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (and age) of the rocks undergoing weathering above that predicted by the idealised trend shown in figure 1a; 2) a decrease in the mantle Sr input and/or seafloor spreading rates; 3) an increase in overall continental weathering rates; and/or 4) an increase in the silicate/carbonate weathering ratio. As I attempt to show below, the most likely explanation involves a combination of options 3 and 4.

The excellent correlation between the normalised seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curve and seawater $\delta^{34}\text{S}$ (Fig. 1b) implies that absolute fluxes and not the isotopic evolution of those

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fluxes led to first-order $^{87}\text{Sr}/^{86}\text{Sr}$ trends. This is because the seawater $\delta^{34}\text{S}$ record is thought to reflect changes in the biogeochemical cycling of sulphur (Strauss, 1999), and so ought to be independent of changes in the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios of rocks exposed to weathering. In this regard, although tectonic upheavals can feasibly change the globally integrated age (Halverson et al., 2007), and therefore $^{87}\text{Sr}/^{86}\text{Sr}$ of rocks undergoing weathering, it seems that such changes only controlled second order variations in the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curve. This conclusion is consistent with the widely held interpretation that the Neoproterozoic-Cambrian interval was a time of CO_2 drawdown (and O_2 increase) caused by elevated weathering rates (Derry et al., 1992). A 50% decrease in the mantle input flux would have been sufficient to cause the Neoproterozoic-Cambrian rise. However, such a large, sustained decrease in seafloor spreading rates seems improbable during a period of exceptionally rapid continental reconfiguration, generally high sea-level and widespread rifting (Kirschvink et al., 1997). The most plausible explanation for the Neoproterozoic-Cambrian rise is therefore a sustained increase in overall continental and/or silicate weathering rates.

Times of low continental influence on ocean $^{87}\text{Sr}/^{86}\text{Sr}$ correlate with the existence of the supercontinents Rodinia and Pangaea, while their break-up is associated with sustained increases in continental influence on the ocean Sr isotope budget. One possible explanation for this connection is that increased rates of physical weathering due to tectonic uplift led to higher overall chemical weathering rates during times of supercontinent break-up and microcontinent collision (Jacobsen and Kaufman, 1999; Squire et al., 2006). In support of this interpretation, the normalised $^{87}\text{Sr}/^{86}\text{Sr}$ curve correlates well with sediment flux rates (Hay et al., 2001) and rifting events (D. S. Bradley, personal communication), while the rise to peak $^{87}\text{Sr}/^{86}\text{Sr}$ values correlates with the formation of huge mountain chains, such as the “Transgondwanan Supermountain” (Squire et al., 2006). However, sustained increases in silicate weathering rates are only thought to be possible so long as increased CO_2 fluxes accompany them (Walker et al., 1981), in which case increased rates of CO_2 degassing from carbonate metamorphism (Bickle, 1994) and granitisation during, may be relevant. Increases in the efficiency of sili-

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cate weathering may also have occurred during the Neoproterozoic-Cambrian interval, caused by the evolution of soil biota (Kennedy et al., 2006; Lenton and Watson, 2004); however, any such irreversible change in the silicate versus carbonate weathering ratio must have been accompanied by a decline in overall continental weathering rates after the Cambrian Period.

It has long been noted that the long-term seawater $^{87}\text{Sr}/^{86}\text{Sr}$ record correlates positively with $\delta^{34}\text{S}$ during the Phanerozoic and negatively with $\delta^{13}\text{C}$ and this has been taken to indicate a tectonic control on global C- and S-biogeochemical cycles (Veizer et al., 1999). The normalised $^{87}\text{Sr}/^{86}\text{Sr}$ curve improves the correlation with $\delta^{34}\text{S}$ and extends it back to c. 800 Ma (Fig. 1b), which implies that increased chemical weathering rates led to higher rates of sulphate reduction (and pyrite burial), presumably linked to higher rates of organic carbon degradation. The alternative proposition that rates of weathering might control seawater $\delta^{34}\text{S}$ directly by changing the riverine sulphate flux can be excluded as this would lead to an inverse correlation between the two isotopic parameters (Bottrell and Newton, 2006). Correlation between globally integrated organic degradation rates and enhanced chemical weathering rates is consistent with the notion that bioproductivity is limited over geological time scales by the availability of phosphorus (Lenton and Watson, 2000), which can only be derived from the weathering of rocks. The onset of the $^{87}\text{Sr}/^{86}\text{Sr}$ rise also marks the beginning of unusually high $\delta^{13}\text{C}$ values that were set to become characteristic of the later Neoproterozoic (Halverson et al., 2007). Thus it would appear that this sustained, weathering-driven increase in productivity also led to higher rates of organic carbon burial, and so presumably was the major driver towards the further oxygenation of the surface environment at this time (Derry et al., 1992).

Recent studies have emphasised the role of terrestrial biota in increasing both P weathering rates (Lenton and Watson, 2004) and overall silicate weathering rates (Kennedy et al., 2006) during the Neoproterozoic-Cambrian interval. Although the evolution of terrestrial eukaryotes by 600 Ma (Heckman et al., 2001; Yuan et al., 2005) may be one cause for the increase in weathering rates, their influence was clearly reversible

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as seawater $^{87}\text{Sr}/^{86}\text{Sr}$ decreased steadily after its Late Cambrian peak. This may be due to the establishment of a new equilibrium between more efficient drawdown of atmospheric CO_2 by silicate weathering and the endogenic CO_2 flux. A similar scenario has been suggested to take place after the evolution of vascular land plants during the later Palaeozoic, whereby the evolution of land plants is considered by some to have led to eutrophication, CO_2 disequilibrium and the contemporaneous Permo-Carboniferous glaciations, and higher atmospheric oxygen levels (Lenton, 2001).

The existence of prolonged pulses of chemical weathering may explain the hitherto enigmatic age distribution of phosphorus-rich rocks on Earth. Sedimentary phosphorite rocks of economic importance have only been deposited during four intervals of Earth history (Shields et al., 2000). The c. 2.0 Ga Paleoproterozoic, Neoproterozoic-late Cambrian (0.65–0.5 Ga) and mid-Cretaceous-Recent (0.1–0.0 Ga) intervals, were true P-giant episodes (Fig. 1b) with phosphorus-rich rocks and related organic-rich strata distributed worldwide. The other major deposit, the Phosphoria Formation, is limited in areal distribution and formed during the Permian icehouse. All three P-giant episodes stand out as times of enhanced chemical weathering rates according to the normalised $^{87}\text{Sr}/^{86}\text{Sr}$ record.

This new reading of the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ record suggests that its first order trend records changes in globally integrated chemical weathering rates, while its second order features are more likely to be related to changes in the isotopic composition of source fluxes (Jones and Jenkyns, 2001). By association, long-term trends in seawater $\delta^{34}\text{S}$ reflect variations in ocean productivity driven by changes in the riverine nutrient flux. Four possible chemical weathering events can be recognised in Earth history, representing periods of disequilibrium and elevated organic productivity. Of these, the Neoproterozoic-Cambrian interval stands out as a time when increased chemical weathering rates pushed the Earth System to a new state characterised by higher O_2 and lower CO_2 and accompanied by the appearance of new forms of animal, plant and fungal life.

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The carbonate dissolution component to river runoff has never been considered in interpretations of first order trends in the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curve despite the dominance of this component today and the certainty that it has must have changed greatly over Earth history. When predictable trends in this parameter and other factors influencing seawater $^{87}\text{Sr}/^{86}\text{Sr}$ are considered, it appears that there was a reversible trend towards unusually high rates of chemical weathering during the late Neoproterozoic and Cambrian. These high rates may help to explain the evidence for eutrophication around this time in the form of massive phosphorite deposits, black shales, high organic carbon burial rates (high $\delta^{13}\text{C}$) and high organic degradation rates (high $\delta^{34}\text{S}$). Sustained high chemical weathering rates and related CO_2 drawdown are both consistent with the notion that Earth's exogenic system became oxygenated over this interval, thus paving the way for the further evolution of animals.

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**A normalised
seawater strontium
isotope curve**

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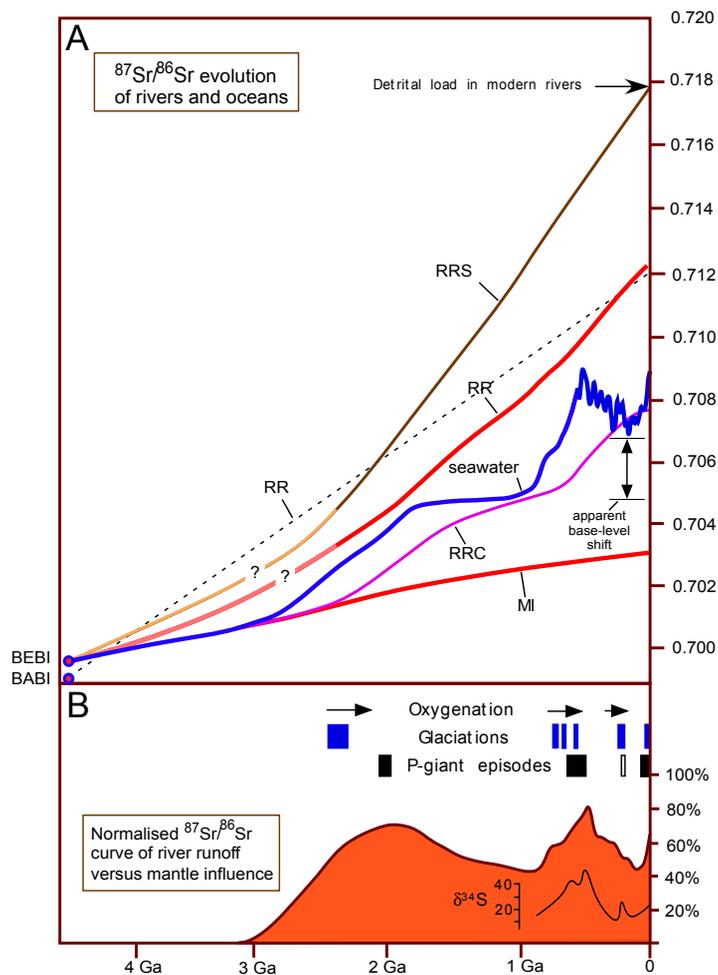
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Fig. 1.

A normalised seawater strontium isotope curve

G. A. Shields

Fig. 1. Sr isotope evolution of seawater and ocean fluxes **(A)** and its implications for the relative influence of continental weathering on ocean composition **(B)**. The seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curve (Veizer et al., 1999; Shields and Veizer, 2002) in (A) has been constructed from the lowermost ratios for each time interval. The river runoff (RR) curve has been determined by assuming a modern-like 9:11 relationship between Sr input from marine carbonate weathering (RRC) and silicate weathering (RRS), respectively. The RRC curve assumes that sediments undergoing weathering have a skewed age distribution, with a mean age of 250 Ma (Peucker-Ehrenbrink and Miller, 2006), and so lags seawater $^{87}\text{Sr}/^{86}\text{Sr}$ by >250 Ma. The RRS curve is an idealistic representation based on predicted crustal evolution (O’Nions et al., 1979), other authors assume much earlier crustal Rb/Sr differentiation with minimal isotopic evolution (Kamber and Webb, 2001). Ocean crust alteration provides less radiogenic Sr to the oceans (MI). The curve in (B) assumes that seawater $^{87}\text{Sr}/^{86}\text{Sr}$ results from simple binary mixing between RR and MI, and shows that the influence of continental weathering was less prior to 2.5 Ga, and negligible prior to 3.0 Ga. The normalised $^{87}\text{Sr}/^{86}\text{Sr}$ and seawater $\delta^{34}\text{S}$ curves (Kampschulte and Strauss, 2004; Hough et al., 2006) have been smoothed by plotting values for every 50 million years only. P-giant episodes (Shields et al., 2000).

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