Modeling the paleo-seawater radiogenic strontium isotope record: A case study of the Late Jurassic-Early Cretaceous

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Abstract

While uniformitarianism underlies many aspects of the Earth sciences, modeling geochemical cycles throughout Earth history requires critically assessing the reliability and constancy of projecting modern fluxes and their isotope ratios into deep geologic time. In this study, we develop an iterative technique for modeling the radiogenic strontium isotope composition ($R_{87} = {}^{87}\text{Sr}/{}^{86}\text{Sr}$) of seawater (sw) during the Phanerozoic utilizing as a constraint the composite record of McArthur et al. (2012) (McArthur, J. M., Howarth, R. J., & Shields, G. A. (2012). Strontium isotope stratigraphy. In F. M. Gradstein, J. G. Ogg, M. Schmitz, & G. Ogg (Eds.), *The Geologic Time Scale 2012* (pp. 127-144): Elsevier). The model yields estimates for the Sr isotope composition of the riverine, submarine groundwater discharge, and diagenetic input fluxes. We use the Late Jurassic-Early Cretaceous segment of the marine Sr isotope record as a case study to demonstrate the efficacy of the new method. To begin, ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ ratios are calculated for a combined riverine and submarine groundwater discharge flux ($tw = \text{total weathering}$) using a prescribed time-variable $\frac{d}{dt}R_{87,sw}$ determined from the LOWESS FIT 5 of McArthur et al. (2012). The calculated $R_{87,tw}$ values are checked against the range of $R_{87,sw}$ for the time interval of interest, as well as older carbonates available for weathering, to determine the reasonableness of the estimates. From these estimates, different prescribed $R_{87,tw}$ values are tested to identify the best match with target seawater ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ ratios and the range of $\frac{d}{dt}R_{87,sw}$. For model runs focusing on long time intervals (≥ 10 Myr), a single prescribed $R_{87,tw}$ value does not produce a reasonable match to the Sr isotope record. Testing the frequency of variation in

$R_{87,tw}$ indicates that during the Early Cretaceous, $R_{87,tw}$ varied at least every 5 Myr, if not more frequently. The minimum frequency at which $R_{87,tw}$ changes likely varied throughout the Phanerozoic, as it depends on the variability of $\frac{d}{dt} R_{87,sw}$ and residence time of Sr. Model results also demonstrate the significance of estimating variation in the total hydrothermal (high and low temperature) input flux of Sr along with seawater Sr concentrations in the geologic past, which are key for properly estimating $R_{87,tw}$

Keywords: LIPs, weathering, hydrothermal, Sr concentration

1. Introduction

The radiogenic strontium isotope composition ($^{87}\text{Sr}/^{86}\text{Sr}$) of seawater represents a balance between Sr inputs from continental weathering (riverine $\pm$ submarine groundwater discharge) with high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and hydrothermal alteration of oceanic crust with low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios relative to seawater (Brass, 1976; Kump, 1989; Palmer and Edmond, 1989; Berner and Rye, 1992; Francois and Walker, 1992; Richter and Turekian, 1993; Davis et al., 2003; Vance et al., 2009; Allegre et al., 2010; Krabbenhoft et al., 2010; Peucker-Ehrenbrink et al., 2010; Beck et al., 2013). The long-term evolution of seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios recorded in marine biogenic calcite have been used to reconstruct changes in the balance between weathering and hydrothermal inputs to the ocean (Brass, 1976; Hodell et al., 1989, 1990; Kump, 1989; Berner and Rye, 1992; Francois and Walker, 1992; Richter and Turekian, 1993; Jones and Jenkyns, 2001; Coogan and Dosso, 2015). Because the two input fluxes are also primary sources for most other elemental cycles (e.g., carbon, sulfur, and iron), quantifying secular changes in the Sr isotope composition of seawater also yields information critical for understanding the long-term evolution of climate

and other Earth surface processes (e.g., Berner and Rye, 1992; Francois and Walker, 1992; Jones and Jenkyns, 2001; Wallmann, 2001, 2004).

Several time-dependent variables control the mass of seawater Sr and its isotope composition. Waltham and Grocke (2006) note that solving the equations (see Section 2) for the marine Sr cycle is “under-constrained rather than unconstrained.” When modeling the Sr isotope composition of seawater, it is critical to identify how the controlling variables might have changed through time. Most model reconstructions of the Phanerozoic Sr isotope record use modern values for the Sr isotope ratio of the riverine, hydrothermal, and diagenetic fluxes, as well as the total mass of Sr in seawater (Jones and Jenkyns, 2001; Wallmann, 2001; Hansen and Wallmann, 2003; Young et al., 2009; Pogge van Strandmann et al., 2013). While measurements of the modern system offer important constraints for interpreting the marine Sr isotope record, allowing the controlling variables to temporally vary should yield more accurate reconstructions.

This paper presents an iterative method for estimating the Sr isotope composition of input fluxes in the geologic past using the composite seawater $^{87}\text{Sr}/^{86}\text{Sr}$ record of McArthur et al. (2012). New model equations that track each Sr isotope individually are employed in this method to avoid the incorrect assumption that the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (i.e., the ratio of two minor isotopes) can account for the total moles of Sr in seawater and the given input fluxes. To begin with, we review estimates for the fluxes and their isotope ratios that control the modern marine Sr cycle and describe how these variables can be constrained when interpreting ancient conditions. Second, we focus on the Late Jurassic to Early Cretaceous (160.5-105.5 Ma) as a case study because the Sr isotope record exhibits a near-constant, linear rise for 30 Myr (which provides a period of stable, consistent pre-conditions for a system that rarely achieves isotopic steady state) prior to experiencing a large downward perturbation, presumably due to the emplacement of

multiple large igneous provinces (LIPs), increased continental arc volcanism, and elevated mid-ocean ridge (MOR) spreading rates (Fig. 1) (Larson, 1991; Jones and Jenkyns, 2001; Rowley, 2002; Hansen and Wallmann, 2003; Bryan and Ernst, 2008; Seton et al., 2009, 2012; McArthur et al., 2012; Bryan and Ferrari, 2013; McKenzie et al., 2016; Wang et al., 2016).

2. Methods

The Supplemental Material provides a complete derivation of the model equations. The model separately tracks each isotope to avoid the assumption that one isotope represents the entire mass of Sr composing seawater or a given flux. For some isotopic systems, such as C, O, and S, where the dominant isotope represents >95 % of the total abundance and/or fractionations are not excessively large, this assumption makes little difference. However, when tracking two minor isotopes, such as ^{87}Sr and ^{86}Sr , where the abundance ratios are much closer to one than <0.05, this assumption can produce small but important differences in the modeled $R_{87,\text{sw}}$ depending on the range of seawater $^{87}\text{Sr}/^{86}\text{Sr}$ being examined. When the range of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios is relatively small, such as might be expected for discrete intervals of geologic time (e.g., the Neogene, Hodell, 1989), assuming ^{86}Sr represents a fixed proportion of total mass of Sr in seawater becomes a reasonable approximation (Richter and Turekian, 1993). However, when modeling longer intervals of time, such as the entire Phanerozoic $\pm$ Neoproterozoic (e.g., Kump, 1989), the range of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios becomes large enough where differences can arise in the model results. The derivation presented in the Supplemental Material first considers the most general case where mass-dependent fractionation of Sr isotopes is a factor (Krabbenhoft et al., 2010; Vollstaedt et al., 2014). In this study, however, we are only concerned with the radiogenic Sr isotope composition of seawater. Therefore, the individual isotope mass-balance equations

simplify to:

$$\frac{d}{dt} {}^{84}\text{Sr}_{sw} = \sum_i \frac{R_{84}}{R_{87,i} + Z} \times F_i - F_{tb} \times \frac{R_{84}}{R_{87,sw} + Z} \quad (1)$$

$$\frac{d}{dt} {}^{86}\text{Sr}_{sw} = \sum_i \frac{1}{R_{87,i} + Z} \times F_i - F_{tb} \times \frac{1}{R_{87,sw} + Z} \quad (2)$$

$$\frac{d}{dt} {}^{87}\text{Sr}_{sw} = \sum_i \frac{R_{87,i}}{R_{87,i} + Z} \times F_i - F_{tb} \times \frac{R_{87,sw}}{R_{87,sw} + Z} \quad (3)$$

$$\frac{d}{dt} {}^{88}\text{Sr}_{sw} = \sum_i \frac{R_{88}}{R_{87,i} + Z} \times F_i - F_{tb} \times \frac{R_{88}}{R_{87,sw} + Z} \quad (4)$$

where $Z = 1 + R_{84} + R_{88}$ is a constant reflecting fixed isotope abundance ratios for ${}^{88}\text{Sr}/{}^{86}\text{Sr}$ ($R_{88} = 8.375209$) and ${}^{84}\text{Sr}/{}^{86}\text{Sr}$ ($R_{84} = 0.056584$), ${}^A N_{sw}$ is the mass of isotopes ${}^{87}\text{Sr}$, ${}^{86}\text{Sr}$, ${}^{88}\text{Sr}$, and ${}^{84}\text{Sr}$ in the oceans, $R_{87,sw}$ is the radiogenic Sr isotope ratio of seawater, and $R_{87,i}$ represents the radiogenic isotope ratio of the hydrothermal (ht), riverine (r), submarine groundwater discharge (sgd), and diagenetic (d) fluxes (F_i), and F_{tb} is the total burial flux. The revised equation for the rate of change of $R_{87,sw}$

$${}^T N_{sw} \frac{d}{dt} R_{87,sw} = \sum_i B (R_{87,i} - R_{87,sw}) F_i \quad (5)$$

where ${}^T N_{sw}$ is the total mass of all Sr isotopes in the oceans, now includes the factor B

$$B = \frac{Z + R_{87,sw}}{Z + R_{87,i}} \quad (6)$$

If mass-dependent fractionation is to be considered, then the term Z would be variable instead of

constant, and fractionation factors for the different burial fluxes would need to be included (see Supplemental Material).

3. Modern Sr cycle fluxes and isotope ratios

3.1 Hydrothermal

The passage of seawater through basaltic oceanic crust at MORs and submarine volcanoes acts as both a sink and source of Sr while yielding extensive Sr isotope exchange between the evolving water and rock (Palmer and Edmond, 1989; Davis et al., 2003; Coogan, 2009; Coogan and Dosso, 2012, 2015). Therefore, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of hydrothermal fluids represents a mixture of Sr leached from basalt ($^{87}\text{Sr}/^{86}\text{Sr} \sim 0.7025$) and seawater. Initial observations and experiments suggested that Sr behaves conservatively during hydrothermal circulation at MOR axial systems (Edmond et al., 1979; Elderfield and Greaves, 1981; Albarede et al., 1981; Berndt et al., 1988; Edmonds and Edmond, 1995; Davis et al., 2003). This would imply that despite significant isotope exchange during water-rock interactions and mineral precipitation, the amount of Sr leached from the oceanic crust equals that removed (see Kump, 1989). However, more recent observations and mass balance modeling of axial hydrothermal systems indicate that axial high temperature hydrothermal systems are a net source of Sr with hydrothermal fluids exhibiting enrichments up to ~ 1.5 times seawater concentration (Barker et al., 2008; Coogan and Dosso, 2012).

Utilizing a simple three-component mixing model, which assumes that Sr behaves conservatively within hydrothermal systems, and further assuming that the modern ocean is in steady-state with respect to Sr concentration and isotope composition, Palmer and Edmond (1989) estimated the modern hydrothermal Sr flux via difference. In their calculation, Palmer

and Edmond (1989) used their estimates for the flux and isotope ratio of the riverine input (see below), the published diagenetic flux and Sr isotope ratio of Elderfield and Gieskes (1982), and an average hydrothermal end-member fluid Sr isotope ratio of 0.7035. These values produced a modern, steady-state, high-temperature hydrothermal Sr flux of 15×10^9 mol/yr, which is ~5-10 times greater than other estimates (Palmer and Edmond, 1989; Butterfield et al., 2001; Davis et al., 2003). The large discrepancy originates from the low riverine flux, which Palmer and Edmond (1989) noted was likely an underestimation, as well as their assumption that the Sr isotope composition of the modern ocean is in steady-state (Vance et al., 2009; Krabbenhoft et al., 2010).

More recently, Davis et al. (2003) attempted to better constrain the hydrothermal Sr input utilizing Sr isotope alteration profiles of oceanic crust and ophiolites. With a direct focus on isotope exchange between seawater and basalt, they recalculated a pure basaltic hydrothermal flux having an $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7025 instead of a total hydrothermal flux, which is a mixture of seawater and basaltic components (i.e., Palmer and Edmond (1989) $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7035). Davis et al. (2003) estimated an axial high temperature hydrothermal basaltic flux of $1.5\text{-}1.8 \times 10^9$ mol/yr. Submarine arc-related hydrothermal systems today are thought to have a total fluid flux equal to ~10 % of the global discharge from MORs, with fluids having much greater concentrations than those from axial systems (Baker et al., 2008; de Ronde et al., 2014 and references within). Davis et al. (2003) included a separate additional arc-related high temperature flux of $\sim 7.0 \times 10^8$ mol/yr. Instead of balancing the marine Sr budget with the low-temperature off-axis hydrothermal flux, Davis et al. (2003) predicted a more modest flux of $\sim 8.0 \times 10^8$ mol/yr assuming 20 % exchange of Sr isotopes over 550 m of oceanic crust. This led to a cumulative hydrothermal basaltic Sr flux estimate of $3.1 \pm 0.8 \times 10^9$ mol/yr (Davis et al., 2003).

Using compilations of alteration mineral assemblages within oceanic sheeted dike complexes and axial hydrothermal fluid compositions Coogan and Dosso (2012) applied a numerical Bayesian inversion procedure to produce internally consistent estimates of fluid and elemental fluxes through axial MOR systems. Utilizing a modern-day oceanic crustal production rate of $2.9 \pm 0.1 \text{ km}^2/\text{yr}$ (Seton et al., 2009), Coogan and Dosso (2012) estimated a global axial fluid flux of $8.0 \pm 2.1 \times 10^{12} \text{ kg/yr}$, which is at the lower end of the range reported in prior studies (Palmer and Edmond, 1989; Elderfield and Shultz, 1996; Davis et al., 2003; Teagle et al., 2003). The Bayesian inversion procedure of Coogan and Dosso (2012) led to an axial hydrothermal Sr flux estimate of $1.30 \pm 0.36 \times 10^9 \text{ mol/yr}$ with an $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.70387. This mixed basaltic-seawater Sr isotope ratio indicates that ~82 % of the Sr is basaltic (0.7025), which corresponds to a basaltic Sr flux of $1.07 \pm 0.30 \times 10^9 \text{ mol/yr}$ (Davis et al., 2003; Coogan and Dosso, 2012). This estimate could be low for axial systems, as it does not consider any water-rock reactions occurring within deep gabbros (Theissan-Krah et al., 2016). The modern low-temperature, off-axis, basaltic hydrothermal flux of Sr today is estimated to be $\leq 2.3 \times 10^9 \text{ mol/yr}$ as diagenetic carbonate precipitation within the altered oceanic crust retains some of the leached Sr (Coogan and Dosso, 2015). Therefore, the modern total basaltic Sr hydrothermal flux is likely $3\text{-}4 \times 10^9 \text{ mol/yr}$.

3.2 Diagenetic

Relative to the hydrothermal and riverine inputs, the diagenetic flux of Sr into the oceans has received less attention because it is thought to represent a minor component of the Sr cycle (Elderfield and Gieskes, 1982; Palmer and Edmond, 1989; Richter and Liang, 1993). Analyses of interstitial waters from 37 Deep Sea Drilling Program (DSDP) sites demonstrate that the concentration and isotope composition of Sr released during carbonate recrystallization controls

the concentration and isotope composition of Sr in these waters (Elderfield and Gieskes, 1982). While some research has suggested that Sr leached from mafic volcanic material dispersed within the sediments might contribute to the diagenetic flux (Elderfield and Gieskes, 1982; Jones et al., 2012a), model calculations assessing the rate of Sr exchange between carbonates and sedimentary porewaters indicates the contribution is minor (Richter and Liang, 1993). Using a compilation of interstitial waters from DSDP cores, Elderfield and Gieskes (1982) estimated the global Sr diagenetic flux and $^{87}\text{Sr}/^{86}\text{Sr}$ ratio to be 3.4×10^9 mol/yr and 0.7084, respectively. Model calculations for Sr exchange during diagenesis indicates ~15 % loss of Sr from seafloor carbonates globally (Richter and Liang, 1993). Assuming Sr burial and riverine fluxes are approximately equal, Richter and Liang (1993) estimated a diagenetic Sr flux of $\sim 5 \times 10^9$ mol/yr with a Sr isotope composition similar to contemporary seawater (max difference 0.0007). Given that the isotopic composition of the diagenetic flux of Sr to the ocean is controlled by recrystallization of previously deposited marine carbonates, it should be noted that the value from Elderfield and Gieskes (1982) matches the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ record at ~20 Ma (McArthur et al., 2012).

3.3 Riverine

Challenges exist to constrain the concentration and $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the modern global dissolved riverine input due to the complexity of lithology, bedrock age, climate, and weathering history within individual drainage basins (Palmer and Edmond, 1989; Vance et al., 2009; Allegre et al., 2010; Krabbenhoft et al., 2010; Peucker-Ehrenbrink et al., 2010; Luo et al., 2014; Voss et al., 2014; Pearce et al., 2015). Riverine Sr concentrations can vary over three orders of magnitude from $<0.1 \mu\text{mol/L}$ to $>100 \mu\text{mol/L}$ (Palmer and Edmond, 1989; Krabbenhoft et al., 2010; Voss et al., 2014; Pearce et al., 2015). Within a single river, Sr concentrations can vary

seasonally from less than a factor of two up to an order of magnitude (Bagard et al., 2011; Luo et al., 2014; Voss et al., 2014; Chapman et al., 2015). The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the global riverine input largely reflects three-component mixing between young mafic rocks (basalt), old felsic rocks (granites), and marine carbonates. In the current icehouse climatic state, riverine Sr concentrations and isotopic ratios may also be impacted by substrate age and variable chemical weathering rates following the retreat of major continental ice sheets and alpine glaciers (Taylor and Blum, 1995; Blum, 1997; Blum and Erel, 1997; Brantley et al., 1998; Millot et al., 2002; White and Brantley, 2003; Vance et al., 2009; von Blanckenburg et al., 2015; Mokadem et al., 2015). A detailed study of the Fraser River Basin, Canada demonstrates the variability and complexity of Sr concentrations and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios within a single large river system (Voss et al., 2014). Within the multiple tributaries and sub-basins, as well as various points along the main stem, Sr concentrations and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios range from 0.126-2.757 $\mu\text{mol/L}$ and 0.70410-0.75150, respectively (Voss et al., 2014). Time series analysis at the mouth of the Fraser River shows a more limited, but still significant range, of values for both parameters, which clearly correlate with seasonal changes (Voss et al., 2014). Voss et al. (2014) ultimately calculated a discharge weighted Sr concentration and $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for the Fraser River of 0.85 ± 0.04 $\mu\text{mol/L}$ and 0.7120 ± 0.0003 , respectively. Other time series and spatial analyses conducted for drainage basins in China, Myanmar, and Siberia have also documented distinct seasonal and spatial variability in Sr concentrations and isotope ratios (Bagard et al., 2011; Wei et al., 2013; Luo et al., 2014; Chapman et al., 2015). These studies highlight the limitations and uncertainty imposed by using compilations of spot measurements to estimate the concentration and isotope ratio of the global riverine input. Along these lines, Palmer and Edmond (1989) noted that most of the river water samples in their compilation were biased towards high to moderate discharge,

implying possible underestimation of the annual Sr flux.

The compilation of riverine Sr concentrations and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios by Palmer and Edmond (1989) comprises eight of the ten largest rivers by discharge, which collectively constitute over 47 % of the total global runoff. However, the authors acknowledged lack of data for several rivers, especially those east of the Ural Mountains. The weighted mean of the data produced a global riverine Sr flux of 3.33×10^{10} mol/yr (average concentration of 0.89 $\mu\text{mol/L}$) with an $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7119. Using more recent data, Davis et al. (2003) calculated similar values of 3.4×10^{10} mol/yr (average concentration of 0.902 $\mu\text{mol/L}$) and 0.7116, respectively. In contrast, the compilation of riverine $^{87}\text{Sr}/^{86}\text{Sr}$ ratios by Allegre et al. (2010) containing more than 30 of the world's principal rivers produced a discharge weighted average $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7136 and a global riverine Sr flux of 1.88×10^{10} mol/yr. With such a low continental riverine flux estimate relative to previous estimates, Allegre et al. (2010) argued for the presence of an additional 4.24×10^{18} g/yr water flux from mafic volcanic islands with a near-mantle $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7035 to address the imbalance. The Sr concentration of this surface water flux from mafic volcanic islands is poorly constrained and could range from 35-200 ppb (Allegre et al., 2010). Combining the minimum (i.e., most conservative) concentration estimate for surface waters from mafic volcanic islands with the continental riverine flux increases the total river flux to 2.05×10^{10} mol/yr, while reducing the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio to 0.7128 (Allegre et al., 2010).

Peucker-Ehrenbrink et al. (2010) developed a model to determine the global riverine Sr flux and $^{87}\text{Sr}/^{86}\text{Sr}$ ratio based on bedrock age and lithology. This model also uses a riverine chemistry database containing 193 rivers representing 50.3 % of the exorheic land surface and 48.3 % of the global exorheic water discharge. The rivers were grouped into 19 large-scale drainage regions comprising non-glaciated land surfaces. Within each drainage region, weighted

Sr concentrations, isotope ratios, and water discharges were determined before extrapolating to the ice-free area of the entire drainage region (Peucker-Ehrenbrink et al., 2010). The model produced a global Sr flux of 4.7×10^{10} mol/yr (average concentration of 1.22 $\mu\text{mol/L}$) and an $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7111 (Peucker-Ehrenbrink et al., 2010). This estimate is the least radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for the global riverine flux available within the literature, which Peucker-Ehrenbrink et al. (2010) attributed to difference in bedrock age calculated according to the Sr and water fluxes – 339 vs. 394 Myr. This difference in bedrock age indicates a greater contribution from younger sedimentary and volcanic rocks with lower $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. It is possible that the impact of younger sedimentary and volcanic rocks on the global riverine $^{87}\text{Sr}/^{86}\text{Sr}$ ratio could be greater than currently estimated given the lack of data for smaller coastal rivers, island arcs, and oceanic islands (Allegre et al., 2010; Peucker-Ehrenbrink et al., 2010).

Beyond the dissolved load, rivers also deliver a significant suspended particulate and bedload to the oceans that can significantly impact global elemental cycles (Meybeck et al., 2003; Syvitski et al., 2003; Gislason et al., 2006; Jones et al., 2012a, b; Jeandel and Oelkers, 2015). Over 60 % of the suspended sediment flux is thought to originate from small mountainous rivers draining geologically young terrains with wet climates (Milliman and Farnsworth, 2011). The majority of such river systems are located within continental volcanic arcs, as well as volcanic and tectonically active islands. Global estimates of the suspended particulate and bedload material delivered to the oceans annually is an order of magnitude greater than the total dissolved load (16.6-30 Gt/yr vs. ~ 1 Gt/yr) (Meybeck et al., 2003; Syvitski et al., 2003; Milliman and Farnsworth, 2011; Jeandel and Oelkers, 2015). Therefore, dissolution of even minor amounts of this sediment flux upon entering seawater could notably contribute to mass and isotope balances of global cycles for such elements as Sr, Nd, Fe, and Si (Gislason et al., 2006; Jones et

al., 2012a, b; Jeandel and Oelkers, 2015). Recent experimental work has highlighted the reactivity of the basaltic component of the sediment flux with regards to Sr, where similar to MOR hydrothermal systems, reaction with seawater causes significant isotopic exchange with only minor changes in concentration (Jones et al., 2012a, b). Using separate experimentally determined mean Sr release rates for continental and volcanic material, as well as assuming 45 % of the riverine particulate flux is volcanically derived, Jones et al. (2012b) conservatively estimated the Sr particulate release flux to be 5.2×10^9 mol/yr. With ~70 % of this flux originating from volcanic sediment the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of this input is likely less radiogenic than seawater.

3.4 Submarine Groundwater Discharge

Similar to the riverine input flux, estimates for the Sr concentration and isotope ratio of submarine groundwater discharge (SGD) are highly variable, as the input determined for any given system is influenced by bedrock age, lithology, history of water-rock interaction within the drainage basin-aquifer system, and exchange during subterranean mixing between fresh and saline groundwaters (Basu et al., 2001; Dowling et al., 2003; Allegre et al., 2010; Beck et al., 2013). Groundwater Sr concentrations can vary over two to three orders of magnitude (0.1-100 $\mu\text{mol/L}$) with higher concentrations (>20 $\mu\text{mol/L}$) typically controlled by mixing with seawater (Basu et al., 2001; Dowling et al., 2003; Beck et al., 2013). In many locations, the riverine and groundwater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are broadly similar; however, the distribution and/or type of rocks composing subterranean aquifer systems can differ from those exposed at the surface (Basu et al., 2001; Dowling et al., 2003; Beck et al., 2013). While drainage basins comprising old granitic and metamorphic rocks, such as the Ganges-Brahmaputra basin, can yield large SGD fluxes, most SGD originates from relatively young coastal carbonate platforms (e.g., Yucatan and

Florida Peninsulas) and volcanic islands (Basu et al., 2001; Allegre et al., 2010; Beck et al., 2013). Thus, estimates for the SGD $^{87}\text{Sr}/^{86}\text{Sr}$ ratio are relatively low (0.7089) (Basu et al., 2001; Beck et al., 2013), which implies that the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the total weathering flux (riverine + SGD) is slightly lower than that for the riverine input. SGD fluxes are highly uncertain. Current estimates range from $7.1\text{-}16.5 \times 10^9$ mol/yr (Basu et al., 2001; Beck et al., 2013).

3.5 Burial Flux and Residence Time

Strontium is removed from the oceans through carbonate and sulfate precipitation and via alteration of the oceanic crust during hydrothermal circulation (e.g., epidote, anhydrite, and carbonate). Direct estimates of Sr burial fluxes are poorly constrained because Sr is a minor-to-trace component within marine carbonates, sulfates, and epidote, requiring assumptions about variable concentrations within each host mineral. Most previous marine Sr models followed initial observations of axial hydrothermal systems and presumed conservative behavior for Sr during high-temperature hydrothermal circulation within the oceanic crust (e.g., Kump, 1989; Davis et al., 2003). In the model equations presented above, the hydrothermal burial flux, either balanced or unbalanced, is embedded in the total burial flux. Because carbonate deposition greatly exceeds deposition of anhydrite, barite, and epidote (both individually and combined), the Sr burial flux is primarily tied to the carbonate burial flux, except during massive basin-scale evaporite (calcium sulfate) deposition (Krabbenhoft et al., 2010; Vollstaedt et al., 2014). Considering the wide variation and uncertainty in estimates for Sr burial within carbonates, most models ignore minor output terms, such as sulfate mineral precipitation, as they would not affect model calculations (Krabbenhoft et al, 2010 and references within; Vollstaedt et al., 2014).

Aragonite incorporates more Sr than calcite because strontianite (SrCO_3) is isostructural with aragonite. Substitution of Sr into CaCO_3 is sensitive to temperature, saturation state, and

growth parameters for biogenic CaCO_3 , as well as the Sr/Ca ratio of seawater, which varies spatially within the modern ocean and throughout geologic time (de Villiers et al, 1994; de Villiers, 1999; Gothmann et al., 2015). Using carbonate burial estimates for different carbonate secreting organisms (i.e., corals, cooccoliths, planktonic foraminifera) and the mean molar Sr/Ca ratio for each group, Krabbenhoft et al. (2010) estimated a modern, total carbonate Sr burial flux of 174×10^9 mol/yr, which is approximately three times higher than estimates for the total input flux of Sr. In contrast, mass-balance model calculations using Phanerozoic radiogenic and stable Sr isotope ratios ($^{87}\text{Sr}/^{86}\text{Sr}$ and $^{88}\text{Sr}/^{86}\text{Sr}$) suggest carbonate Sr burial fluxes range from $0\text{--}47 \times 10^9$ mol/yr, with a mean value of 16×10^9 mol/yr, which is approximately half to a third of the riverine flux (Palmer and Edmond, 1989; Peucker-Ehrenbrink et al., 2010; Vollstaedt et al., 2014). Given the degree of uncertainty surrounding Sr burial flux estimates, as well as the lack of impact on seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, the Sr burial flux is often ignored or substituted with assumptions about a constant residence time (Hodell et al., 1989, 1990; Jones and Jenkyns, 2001; Wallmann, 2001; Hansen and Wallmann, 2003; Walthman and Grocke, 2006; Young et al., 2009; Pogge van Strandmann et al., 2013). The modern residence time for seawater Sr – calculated by dividing the mass of Sr in seawater by the total input flux – is estimated at ~ 2 Myr (Hodell et al., 1989, 1990; Palmer and Edmond, 1989; Basu et al., 2001; Peucker-Ehrenbrink et al., 2010; Beck et al., 2013). Assuming a constant residence time, which presumes total input and output fluxes are in steady-state, limits concentration changes during major perturbations and artificially returns concentrations to initial values following the end of perturbations.

4. Estimating Sr cycle fluxes and isotope ratios for the Late Jurassic-Early Cretaceous

The global riverine (dissolved and particulate) and SGD $^{87}\text{Sr}/^{86}\text{Sr}$ ratios likely varied

throughout the Phanerozoic given variations in the type and age of bedrock exposed to weathering (Wallmann, 2004; Peucker-Ehrenbrink et al., 2010; Beck et al., 2013; Voss et al., 2014), as well as the time dependent nature of weathering reactions (Taylor and Blum, 1995; Blum, 1997; Blum and Erel, 1997; Brantley et al., 1998; White and Brantley, 2003). Therefore, assuming that ancient Sr inputs have the same isotopic composition as modern inputs introduces uncertainty into model calculations. Because the complete distribution and age of marine carbonates, felsic rocks, and mafic rocks available for weathering on the continents at any time during the deep geologic past are unknown, the isotopic composition of the total weathering flux is a poorly constrained parameter (Equations 1-5). In contrast, Sr isotope analyses of microfossils, shell material, and bulk carbonate from ODP cores and land based samples over the past 25 years have greatly improved the density of Phanerozoic $^{87}\text{Sr}/^{86}\text{Sr}$ data and allowed construction of a high-resolution, composite seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curve (McArthur et al., 2012 and references within). Here, we suggest that the LOWESS FIT 5 of McArthur et al. (2012) (with time steps of 50 kyr) provides a key constraint to Equation 5 that has been underutilized, namely the rate of change of seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios ($\frac{d}{dt}R_{87,sw}$). The method for modeling the marine Sr cycle presented below is an iterative process that begins with calculating an $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for a combined riverine (dissolved and particulate) and SGD flux ($F_{tw} = F_r + F_{sgd}$, tw = total weathering) that produces a reasonable match with the McArthur et al. (2012) $^{87}\text{Sr}/^{86}\text{Sr}$ record given prescribed values for the other parameters.

In the first step of this iterative process a set of $\frac{d}{dt}R_{87,sw}$ values is determined from the LOWESS FIT 5 of McArthur et al. (2012). Selection of the $\frac{d}{dt}R_{87,sw}$ time-step depends on the magnitude and frequency of the inflections within the Sr isotope record, along with the need to match each inflection within the composite curve. For this case study, with a time interval of

interest spanning 55 Myr (160.5-105.5 Ma), constant $\frac{d}{dt}R_{87,sw}$ values were determined for intervals of varying length ranging from 0.1-2.4 Myr (Supplemental Table 2). This set of $\frac{d}{dt}R_{87,sw}$ is necessary to calculate an array of $R_{87,tw}$ according to

$$R_{87,tw} = \frac{Z \left(\frac{{}^T N_{sw} \times \frac{d}{dt} R_{87,sw} - B(R_{87,ht} - R_{87,sw})F_{ht} - B(R_{87,d} - R_{87,sw})F_d}{F_{tw}} \right) + R_{87,sw} \times (Z + R_{87,sw})}{Z + R_{87,sw} - \left(\frac{{}^T N_{sw} \times \frac{d}{dt} R_{87,sw} - B(R_{87,ht} - R_{87,sw})F_{ht} - B(R_{87,d} - R_{87,sw})F_d}{F_{tw}} \right)} \quad (7)$$

Equations 1-4 (modified for $F_{tw} = F_r + F_{sgd}$) are solved simultaneously to determine $R_{87,sw}$ using the initial resultant $R_{87,tw}$ from Equation 7 to enable calculation of $R_{87,tw}$ at the next time step. Multiple arrays of $R_{87,tw}$ are calculated using a range of different initial input fluxes, perturbation factors, and seawater Sr concentrations.

A measure of the strength of the hydrothermal or total weathering levers in the marine Sr cycle can be assessed by the difference between the R_{87} of the input flux and seawater.

Therefore, the resulting arrays of $R_{87,tw}$ from Equation 7 are checked against the composite seawater Sr isotope record of McArthur et al. (2012) for the time interval of interest to evaluate the potential strength of the hydrothermal and total weathering levers. In addition, the sets of $R_{87,tw}$ are compared to older portions of the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ record to assess the potential availability of marine carbonates exposed on the continents for weathering that would reasonably contribute to a three-component mixture producing values within this range. Following these two assessments, the variability within the most reasonable arrays of calculated $R_{87,tw}$ guides the

estimation of a set of prescribed $R_{87,tw}$ with predetermined time intervals. Using Equations 1-4 (modified for $F_{tw} = F_r + F_{sgd}$) the set of prescribed $R_{87,tw}$ is tested and modified to produce the most reasonable overall match with target seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (i.e., inflection points and plateaus) and the range of $\frac{d}{dt} R_{87,sw}$ determined from the LOWESS FIT 5 of McArthur et al. (2012). Table 1 presents the fluxes, isotope ratios, oceanic concentrations, and perturbation factors used for the Late Jurassic-Early Cretaceous marine Sr cycle model calculations presented below.

4.1 Hydrothermal

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the hydrothermal flux depends whether a purely basaltic hydrothermal flux or a total hydrothermal flux is assumed (Palmer and Edmond, 1989; Davis et al., 2003; Coogan and Dosso, 2012). Unaltered MOR basalt from the Cretaceous indicates a basaltic hydrothermal $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.7030, which is slightly greater than the modern value of 0.7025 (Coogan, 2009). Model calculations and experiments indicate that basalt-seawater reactions, such as those involving Sr leaching and isotopic exchange, are strongly temperature-dependent at relatively low temperatures (Coogan and Dosso, 2015). A nearly five-fold difference in the carbonate content of altered oceanic crust between the Late Cretaceous and Late Cenozoic that parallels apparent changes in seawater bottom temperatures may express this behavior (Gillis and Coogan, 2011; Coogan and Gillis, 2013; Coogan and Dosso, 2015). Therefore, the 10-15°C increase in bottom water temperature estimated for the Cretaceous compared to the present would produce a four- to five-fold increase in the low-temperature, off axis hydrothermal Sr flux (Coogan and Dosso, 2015).

Relatively higher MOR spreading rates during the Cretaceous due to the continued break up of Pangea would have impacted both the low-temperature off axis and high-temperature axial

hydrothermal fluxes (Larson, 1991; Rowley, 2002; Seton et al., 2009, 2012). Crustal production rates during the Cretaceous were 1.38 ± 0.18 times higher than present (Seton et al., 2009). Therefore, the Cretaceous low-temperature off axis hydrothermal Sr flux, impacted by both elevated bottom water temperatures and increased MOR spreading rates, would have been $12.4 \pm 2.1 \times 10^9$ mol/yr if the modern flux is presumed to be $\sim 2 \times 10^9$ mol/yr (Seton et al., 2009; Coogan and Dosso, 2015). The initial reactions during recharge flow for the axial, high-temperature hydrothermal Sr flux are also thought to be temperature dependent (Coogan and Dosso, 2015). Assuming temperature-dependent reactions affect the first 10% of the recharge flow, combined with the effect of elevated MOR spreading rates, we estimate an axial, high-temperature hydrothermal basaltic Sr flux of $3.0 \pm 0.2 \times 10^9$ mol/yr during the Cretaceous (Davis et al., 2003; Seton et al., 2009; Coogan and Dosso, 2012, 2015). We estimate that the arc-related hydrothermal Sr flux during the Cretaceous was 1×10^9 mol/yr based on observations that the fluid flux through modern submarine arc-related hydrothermal systems equates to $\sim 10\%$ of the global MOR flux and that Sr concentrations are greater than those for axial (Davis et al., 2003; Barker et al., 2008; de Ronde et al., 2014 and references within). For simplicity, we assume bottom seawater temperatures remain consistently elevated and consider a single, total hydrothermal basaltic Sr flux of $16.4 \pm 2.1 \times 10^9$ mol/yr for the Late Jurassic-Early Cretaceous (Models A-F, Table 1).

Comparing Sr isotope profiles through recently altered oceanic crust to those from Cretaceous ophiolites indicates a higher degree of alteration and transfer of seawater Sr occurred within the sheeted dike complex during the Cretaceous (Davis et al., 2003; Coogan, 2009). Mass balance calculations, which assume that all the seawater Sr reaching the sheeted dike complex entered the crust, yield minimum seawater Sr concentration estimates of $434 \mu\text{mol/kg}$ during the

Cretaceous and 32 $\mu\text{mol/kg}$ for the present (Coogan, 2009). The modern seawater Sr concentration is 90 $\mu\text{mol/L}$ (Pilson, 1998). These estimates are minimums because some seawater Sr is removed during anhydrite precipitation prior to reaching the sheeted dike complex and not all Sr that reaches the sheeted dike complex enters the crust (Davis et al., 2003; Coogan, 2009; Coogan and Dosso, 2012). These minimum estimates correspond to a minimum Sr burial flux of $4.8 \pm 1.3 \times 10^9$ mol/yr via axial high-temperature hydrothermal circulation during the Cretaceous, which is ~18.5X greater than present (Coogan, 2009; Seton et al., 2009; Coogan and Dosso, 2012). In contrast to the present, this implies that axial high temperature hydrothermal circulation was a net sink instead of a net source during the Cretaceous (Barker et al., 2008; Coogan and Dosso, 2012). Determining if axial high temperature hydrothermal circulation is a net source or sink appears to depend on seawater Sr concentrations. Because the seawater Sr concentrations reported in Coogan (2009) are minimums, we assume an initial Sr concentration of 450 $\mu\text{mol/kg}$ for our models, which is consistent with elevated Ca concentrations, as well as a peak in seawater Sr/Ca ratios during this time (Lowenstein et al., 2003; Gothmann et al., 2015). To test the validity of the elevated Sr concentrations in the Cretaceous ocean and the temperature dependence of the hydrothermal flux, models employing a modern Sr concentration (90 $\mu\text{mol/L}$) and hydrothermal fluxes elevated only to account for increased MOR spreading rates, are run for comparison (Models A modern & B modern, Table 1).

4.2 Diagenetic

Considering that the diagenetic flux is a minor component of the Sr cycle and noting that the two estimates for the modern are about the same, the flux from Elderfield and Gieskes (1982) is used for all Late Jurassic-Early Cretaceous models (Table 1). However, the isotope composition of the diagenetic flux, which is controlled by recrystallization of carbonate

sediments present on the seafloor, has varied through time given temporal changes in the isotope composition of seawater (Elderfield and Gieskes, 1982; Richter and Liang, 1993; McArthur et al., 2012; Coogan and Dosso, 2015). Here, we use the LOWESS FIT 5 of McArthur et al. (2012), which provides seawater/carbonate $^{87}\text{Sr}/^{86}\text{Sr}$ ratios with a time step of 50 kyr for most of the Phanerozoic, to provide a more accurate and time variable isotope composition for the diagenetic flux. Following the observation that the modern diagenetic $^{87}\text{Sr}/^{86}\text{Sr}$ ratio corresponds with the Phanerozoic seawater record at 20 Ma, we assume that the isotope composition of the diagenetic input matches the seawater curve at ~20 Myr prior to the time period being modeled. In addition, we allow the value to change every 2.5 Myr. Therefore, we take the average over a 2.5 Myr interval ($n = 51$) centered at 20 Myr prior to model age. Supplemental Table 3 provides the values used for the Sr isotopic composition of the diagenetic flux in all models. These diagenetic $^{87}\text{Sr}/^{86}\text{Sr}$ ratios range from both greater than and less than the corresponding contemporaneous modeled seawater Sr isotope composition, with differences <0.0005 .

4.3 Burial

As noted previously, most models of the marine Sr cycle assume that the burial flux goes as the mass of seawater Sr divided by the modern residence time of ~2 Myr. However, the relatively high Sr concentrations suspected for the Cretaceous (e.g., Coogan, 2009; Gothman et al., 2015) inherently imply a longer residence time. The moderate to low magnitude of $\frac{d}{dt}R_{87,sw}$ ($<3 \times 10^{-5}$, ~83% of the time) during the Late Jurassic-Early Cretaceous compared to the higher rates of change ($>5 \times 10^{-5}$, ~34% of the time) over the last 35 Myr further support higher Sr concentrations with a longer residence time than present (McArthur et al., 2012). Considering the large uncertainty in Sr burial flux estimates we allow the residence time to vary and assume Sr concentrations remain constant except during major perturbations, such as the emplacement of

large igneous provinces (LIPs) and deposition of massive evaporites, when significant concentration changes are expected.

4.4 Riverine and SGD (Total Weathering)

In the initial model runs, where $R_{87,tw}$ is the calculated unknown, the models are paired (e.g., models A and B) to have a low and high F_{tw} (Table 1). In models E and F, F_{tw} represents the high and low dissolved riverine flux estimates, respectively, available in the literature with no contribution from SGD or particulate release from the sediment flux (Palmer and Edmond, 1989; Davis et al., 2003; Peucker-Ehrenbrink et al., 2010). In all other models, F_{tw} is a summation covering the range of estimates for the dissolved riverine, SGD, and particulate release fluxes. The low F_{tw} estimate (47×10^9 mol/yr) is based on values from Palmer and Edmond (1989), Beck et al. (2013), and Jones et al. (2012b), while the high F_{tw} estimate (62×10^9 mol/yr) is based on values from Peucker-Ehrenbrink et al. (2010), Basu et al. (2001), and Jones et al. (2012b). For 30 Myr prior to the Early Cretaceous (Barremian), the marine $^{87}\text{Sr}/^{86}\text{Sr}$ record shows a near constant, linear increase that is assumed to represent stable, consistent preconditions (instead of steady state). It is presumed this increase results from minor, gradual changes in the total weathering flux and/or $R_{87,tw}$. In model runs where $R_{87,tw}$ is being calculated, F_{tw} is assumed to be constant during the 30 Myr increase in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. In the paired model runs where $R_{87,tw}$ values are prescribed, F_{tw} begins at intermediate values (52×10^9 mol/yr or 37×10^9 mol/yr) and gradually increases by 1×10^9 mol/yr every 2.5 Myr over the 30 Myr time interval (Table 2). This gradual increase in F_{tw} , <2 % for models AB and CD and <3 % for model EF, is chosen to bridge the low and high estimates for F_{tw} and reduce the change in $R_{87,tw}$ needed to reproduce the precondition interval.

4.5 Perturbations

Major geologic events during the Early Cretaceous that could broadly impact the marine Sr cycle include emplacement of multiple LIPs, increased continental arc volcanism, elevated MOR spreading rates, and deposition of the anhydrite-rich Late Aptian South Atlantic Salts (Larson, 1991; Jones and Jenkyns, 2001; Rowley, 2002; Hansen and Wallmann, 2003; Davison, 2007; Bryan and Ernst, 2008; Seton et al., 2009, 2012; Chandler et al., 2012; McArthur et al., 2012; Bryan and Ferrari, 2013; Chaboureaud et al., 2013; Hochmuth et al., 2015; McKenzie et al., 2016; Wang et al., 2016). Marine $^{87}\text{Sr}/^{86}\text{Sr}$ ratios increase for 30 Myr reaching a peak during the Barremian (Fig. 1) (McArthur et al., 2012). The significant decrease in $\frac{d}{dt} R_{87,sw}$ leading to the peak and inflection in the Sr isotope record during the Barremian is thought to result from the initial phase of emplacement of the Ontong Java-Manihiki-Hikurangi Plateau (OJMHP), the largest of the Early Cretaceous LIPs (Jones and Jenkyns, 2001; Bryan and Ernst, 2008; Chandler et al., 2012; Bryan and Ferrari, 2013; Hochmuth et al., 2015). The initial decrease in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios during the Barremian-Aptian likely corresponds to the main phase of OJMHP emplacement, while the continued decrease may represent a new, quasi steady-state (mass balance but isotope imbalance) with elevated hydrothermal and total weathering fluxes related to the subsequent emplacement of the Kerguelen Plateau, increased continental arc volcanism, and higher MOR spreading rates (Tables 1 and 2) (Larson, 1991; Rowley, 2002; Bryan and Ernst, 2008; Seton et al., 2009, 2012; Bryan and Ferrari, 2013; McKenzie et al., 2016; Wang et al., 2016). While the deposition of the Late Aptian South Atlantic Salts would not directly impact seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, Sr concentrations would have decreased significantly. While no specific major geologic event appears to correspond with the Late Aptian-Early Albian rise in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, in general, increases in the marine Sr isotope record are presumed to reflect increased riverine inputs and/or decreased hydrothermal inputs (Palmer and Edmond, 1989;

Jones and Jenkyns, 2001; Hansen and Wallmann, 2003; McArthur et al., 2012; Coogan and Dosso, 2015).

According to Equation 7, major unbalanced perturbations to the marine Sr cycle, such as the emplacement of OJMHP, could yield unreasonably large step changes to $R_{87,tw}$ (Fig. 2A). Therefore, $R_{87,tw}$ is assumed to change at a constant, linear rate ($2.0\text{e-}5/\text{Myr}$) during the perturbation instead of being calculated, as they are during the other portions of the model runs. The constant linear rate of change in $R_{87,tw}$ is estimated based on the overall rate of change in $R_{87,tw}$ determined for the 30 Myr precondition interval. Using a prescribed constant, linear rate of change for $R_{87,tw}$ during the OJMHP perturbation while calculating $R_{87,tw}$ during other parts of the model, allows hydrothermal and total weathering fluxes to be adjusted until target seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are met at the end of the initial and main phases of OJMHP (Fig. 2). Once appropriate perturbation fluxes are identified, the linearized rate of change for $R_{87,tw}$ during the OJMHP perturbation is removed and $R_{87,tw}$ values are calculated over the entire model. Modeling attempts to identify appropriate perturbation factors prescribing a $90\text{ }\mu\text{mol/L}$ seawater Sr concentration and elevated total hydrothermal flux ($16.4 \times 10^9\text{ mol/yr}$) produced unrealistically low values given the size of OJMHP. This is consistent with the moderate to low $\frac{d}{dt} R_{87,sw}$ values during such a large perturbation, which indicate a significantly larger reservoir of Sr than present (Coogan, 2009; McArthur et al., 2012). Therefore, such a model was not evaluated further.

5. Model Implementation, Results, and Discussion

5.1 Checking calculated R_{tw} and Sr concentrations

Figure 3 presents the arrays of calculated $R_{87,tw}$ for all eight models listed in Table 1. The difference between the calculated $R_{87,tw}$ and the range of Late Jurassic-Early Cretaceous seawater

$^{87}\text{Sr}/^{86}\text{Sr}$ ratios for models with the higher hydrothermal flux and elevated Sr concentration is similar to that between estimates for $R_{87,r}$ and seawater today (~ 0.002) (Peucker-Ehrenbrink et al., 2010). In contrast, the difference between calculated $R_{87,tw}$ and the range of Late Jurassic-Early Cretaceous seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for models with the lower hydrothermal flux and modern Sr concentration is small (approximately one eighth to one quarter of today's difference). With only two primary levers controlling changes in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, such small differences between the Sr isotope ratio of one lever and seawater will significantly weaken the impact of the other lever. The more reasonable calculated $R_{87,tw}$ for models A-F supports the temperature dependence of the hydrothermal Sr flux and highlights the significance of the low temperature, off axis hydrothermal Sr flux (Coogan and Dosso, 2015). The results also demonstrate that the magnitudes of the total weathering and total hydrothermal fluxes, as well as the seawater Sr reservoir size, are critical for determining reasonable $R_{87,tw}$ in the geologic past.

In Figure 4, the total range of calculated $R_{87,tw}$ values for a 450 $\mu\text{mol/L}$ (Models A-F) and a 90 $\mu\text{mol/L}$ (Models A modern and B modern) Sr ocean are highlighted in light blue and yellow, respectively, against the known Phanerozoic and Neoproterozoic seawater $^{87}\text{Sr}/^{86}\text{Sr}$ record (McArthur et al., 2012). In the three-component mixing model controlling $R_{87,tw}$, a small contribution from granite weathering can have a significant impact. Because granite weathering increases $R_{87,tw}$, the majority of marine carbonates available for weathering should therefore have $^{87}\text{Sr}/^{86}\text{Sr}$ ratios less than or similar to the calculated $R_{87,tw}$ value. However, a large contribution of non-radiogenic Sr from basalt weathering can greatly extend the upper range of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios available from carbonate weathering. For a 90 $\mu\text{mol/L}$ Sr ocean experiencing minimal contributions from basalt weathering, few carbonate rocks older than 160.5 Ma are suitable (below yellow rectangle in Fig. 4). In contrast, a 450 $\mu\text{mol/L}$ Sr ocean that similarly experiences

minimal contributions from basalt weathering allows for a much larger and reasonable range of older marine carbonates available for weathering (below blue rectangle in Fig. 4). The lack of suitable carbonates to weather plus the difference between $R_{87,tw}$ and seawater Sr isotope ratios support the existence of a $\sim 450 \mu\text{mol/L}$ Sr ocean during the Cretaceous, which is consistent with elevated Ca concentrations at this time (Lowenstein et al., 2003; Coogan, 2009; Vollstaedt et al., 2014). In addition, these results indicate the need to include a low temperature, off-axis hydrothermal Sr flux that exhibits a significant temperature dependence in global marine Sr cycle models (Coogan and Dosso, 2015).

5.2 Prescribing constant $R_{87,tw}$ values

Prescribing a single constant $R_{87,tw}$ value for models with long time frames (e.g., 55 Myr), even when appropriately estimated for the time interval of interest, produces $R_{87,sw}$ values and a curve structure that poorly match the McArthur et al. (2012) record (Fig. 5). This result is expected as the iterative model presented here assumes that the global, integrated $R_{87,tw}$ value varies considerably over geologic timescales. Prescribing $R_{87,tw}$ values that change every 10 Myr moderately improves the match to the near-linear Late Jurassic-Early Cretaceous rise in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, including the peak during the initial phase of OJMHP in the Barremian (blue line Fig. 6A and Supplemental Table 4). However, during the main phase of OJMHP, the modeled $R_{87,sw}$ decreases at a steeper rate than the McArthur curve before drastically diverging from the continued Barremian-Aptian decrease following the end of the main phase of the OJMHP eruption (blue line Fig. 6A). Adjusting $R_{87,tw}$ values every 5 Myr improves the match (purple line Fig. 6A and Supplemental Table 4). Adjusting $R_{87,tw}$ values every 2.5 Myr further improves agreement between measured and model $R_{87,sw}$ (red line Fig. 6A and Supplemental Table 4). However, notable differences still remain in the $\frac{d}{dt}R_{87,sw}$ during the main phase of OJMHP, as

well as $R_{87,sw}$ values determined for the Late Aptian minimum and Early-Middle Albian plateau.

The best overall fit with $\frac{d}{dt}R_{87,sw}$ and target seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios occurs when $R_{87,tw}$ is allowed to vary every 2.5 to 1 Myr (Fig. 6 and Table 3). The more frequent variation in $R_{87,tw}$ values is needed during the major perturbations and transitions (inflection points) within the seawater $^{87}\text{Sr}/^{86}\text{Sr}$ record (Tables 2 and 3). This frequency of change is reasonable considering the range of estimates for average denudation rates implying that erosion can remove 10-1000 meters of rock within 1 Myr (Portenga and Bierman, 2011; Herman et al., 2013). In addition, cyclic variation in orbital parameters can influence climate change on timescales ≤ 1 Myr, such as the distribution of precipitation (wet versus dry regions), that could impact the global integrated $R_{87,tw}$ value (i.e., Elkibbi and Rial, 2001; Graziano and Raspini, 2015; Charbonnier et al., 2016).

Although the model presented here presumes the global integrated $R_{87,tw}$ value can vary on geologically short time scales ($\sim 10^6$ yr), the general background level changes in $R_{87,tw}$ are not expected to be large. While the new exposure of a Precambrian granite or large amounts of basalt, for example, could greatly shift river water $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in a single drainage basin, the impact on global $R_{87,tw}$ values is likely to be muted by opposing changes in other drainage basins, the integration of all rivers, and additional contributions from SGD. On the other hand, major orogenic events and the development of new watersheds could drive perturbations in the radiogenic Sr isotope record through large changes in the global integrated $R_{87,tw}$. Therefore, when prescribing $R_{87,tw}$ values during periods without major orogenic events, such as examined in this paper, a goal should be to minimize the magnitude of step changes. The improved match between measured and modeled data obtained by increasing the frequency at which $R_{87,tw}$ changes (Fig. 6A) supports this conclusion. When $R_{87,tw}$ values change every 5 Myr, the magnitude of change is >0.0001 for 50 % of the time compared to only 21 % of the time when

the $R_{87,tw}$ varies every 2.5 to 1 Myr (model AB).

The persistence of large changes in $R_{87,tw}$ values for the “best fit” scenario (Table 3) is interpreted as a need to reassess the flux values. Excluding the precondition interval, there are three large changes in $R_{87,tw}$ that occur near the beginning and end of the main phase of OJMHP, as well as during the Late Aptian-Early Albian rise in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (Table 3 – model times 32, 36 and 45.5 Myr, respectively). Minor adjustments of $\leq 5\%$ to the main phase hydrothermal and weathering perturbation factors, along with 0.15X and 0.20X increases to the post-OJMHP hydrothermal flux in models AB and CD/EF, respectively, significantly reduce the changes in $R_{87,tw}$ and greatly improve the match between modeled and measured $\frac{d}{dt}R_{87,sw}$ (Tables 2-4). The magnitude of the decrease in the hydrothermal flux during the Late Aptian minimum is increased in all models and becomes the dominant mechanism driving the Late Aptian-Early Albian rise in seawater Sr isotope ratios. The change in the total weathering flux at this time is reduced to 5 % in models AB and CD, and 7.5 % in model EF (Tables 2 and 4). During the Albian plateau in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, a 5 % decrease in the total weathering flux is added at model time 50.5 Myr in all models to help minimize changes in $R_{87,tw}$ (Tables 2-4). These adjustments in the fluxes significantly reduce the maximum step size, along with the average and standard deviation of the step size (Table 3 and Fig. 7).

6. Conclusions

This study presents a new, iterative method for modeling the marine Sr cycle in the geologic past. The model capitalizes on the LOWESS FIT 5 of McArthur et al. (2012), which provides constraints and improves estimates for the Sr isotope composition of the input fluxes. The results from a case study focusing on the Late Jurassic-Early Cretaceous time period, show

the importance of estimating variability in the total hydrothermal flux along with seawater Sr concentrations in the geologic past. Results further indicate that the Sr isotope composition of the total weathering input ($R_{87,tw}$) has varied throughout Earth history on short and long geologic timescales. Model results indicate that $R_{87,tw}$ values should be adjusted every 5 Myr, if not more frequently, at least when examining the Early Cretaceous Sr cycle. The minimum frequency at which $R_{87,tw}$ values should be changed can vary throughout the Phanerozoic depending on the rate of change of the seawater Sr isotope record ($\frac{d}{dt} R_{87,sw}$) and the residence time of Sr during the specific time interval being examined. By attempting to match measured $\frac{d}{dt} R_{87,sw}$ and minimize the step size for prescribed $R_{87,tw}$ values, the iterative model also improves flux estimates. The model output supports estimates for a ~450 $\mu\text{mol/L}$ Sr ocean during the Cretaceous and demonstrates that increases in both hydrothermal and total weathering fluxes during the emplacement of OJMHP are needed to produce the Barremian-Aptian decrease in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. Moreover, variations in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios resulting from perturbations to the weathering system are not limited to changes in the flux but could result from shifts in $R_{87,tw}$ while F_{tw} remains nearly constant. The improved constraints and more representative model of the Sr cycle within the geologic past presented here should enhance coupled modeling of linked key biogeochemical cycles (e.g., Sr with C, S, Fe) allowing for better paleoclimatic and paleoceanographic reconstructions.

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Figure Captions

Figure 1. The Late Jurassic-Early Cretaceous seawater $^{87}\text{Sr}/^{86}\text{Sr}$ curve from McArthur et al. (2012) with GTS2012 stage boundaries.

Figure 2. Modeled total weathering (A) and seawater (B) $^{87}\text{Sr}/^{86}\text{Sr}$ ratios demonstrating the use of a prescribed linearized $R_{87,tw}$ value to constrain fluxes and minimize step changes in $R_{87,tw}$ during the emplacement of the OJMHP. The non-linear $R_{87,tw}$ curve (A, black) shows the large theoretical step change in $R_{87,tw}$ for initial unconstrained perturbation fluxes necessary to match the McArthur et al. (2012) curve (B, black). The linear $R_{87,tw}$ and seawater curves (light blue) demonstrate the large step change in $R_{87,tw}$ following the end of the perturbation and the extreme mismatch in modeled $R_{87,sw}$ values and the McArthur et al. (2012) curve upon first linearization of the initial unconstrained perturbation fluxes. Modification to the perturbation fluxes (linear adjusted curves – purple/magenta) eventually results in minimizing the step change in $R_{87,tw}$ following the end of the perturbation while also meeting target seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios at the end of the initial and main phases of the OJMHP perturbation.

Figure 3. Calculated $R_{87,tw}$ values during the Late Jurassic-Early Cretaceous for a 450 $\mu\text{mol/L}$ Sr ocean (Models A-F) and a 90 $\mu\text{mol/L}$ Sr ocean (Models A modern + B modern). Range of seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for Late Jurassic-Early Cretaceous from McArthur et al. (2012) highlighted in gray box. Modern riverine $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (dashed green line) shown for comparison is taken from Peucker-Ehrenbrink et al. (2010).

Figure 4. The Phanerozoic and Neoproterozoic seawater $^{87}\text{Sr}/^{86}\text{Sr}$ record from McArthur et al. (2012) with the total range of calculated $R_{87,tw}$ values highlighted for a 450 $\mu\text{mol/L}$ (light blue) and 90 $\mu\text{mol/L}$ (yellow) Sr ocean during the Late Jurassic-Early Cretaceous.

Figure 5. Modeled seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from models AB, CD, EF, and AB modern with a single, prescribed constant $R_{87,tw}$ value and the McArthur et al. (2012) curve shown for comparison. $R_{87,tw}$ value for models AB and CD is 0.7087. $R_{87,tw}$ value for model EF is 0.7092. $R_{87,tw}$ value for model AB modern is the global riverine estimate of 0.7111 from Peucker-Ehrenbrink et al. (2010).

Figure 6. A) Modeled seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from model AB with prescribed constant $R_{87,tw}$ values varying every 10 Myr, 5 Myr, 2.5 Myr, and a mixture of 2.5 and 1 Myr. McArthur et al. (2012) curve included for comparison. See Supplemental Table 4 for lists of prescribed $R_{87,tw}$ values. B) Modeled seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from models AB, CD and EF with prescribed constant $R_{87,tw}$ values varying a mixture of every 2.5 or 1 Myr. McArthur et al. (2012) curve included for comparison. See Table 3 for list of prescribed $R_{87,tw}$ values for the mixed 2.5 and 1 Myr model runs for all three models.

Figure 7. Calculated $R_{87,tw}$ values for models A and B, same as in Figure 4, along with initial and final prescribed $R_{87,tw}$ for model AB (Table 3). Range of seawater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for Late

Jurassic-Early Cretaceous from McArthur et al. (2012) highlighted in gray box. Modern riverine $^{87}\text{Sr}/^{86}\text{Sr}$ value (dashed green line) for comparison is taken from Peucker-Ehrenbrink et al. (2010).

Table 1. Model values for Late Jurassic-Early Cretaceous Sr cycle model where $R_{87,tw}$ is calculated.

	Model A	Model B	Model C	Model D	Model E	Model F	Model A modern	Model B modern
[Sr] ($\mu\text{mol/L}$)	450	450	450	450	450	450	90	90
$R_{87,sw}$ initial	0.706849	0.706849	0.706849	0.706849	0.706849	0.706849	0.706849	0.706849
F_{tw} (mol/yr)	62e9	47e9	62e9	47e9	47e9	34e9	62e9	47e9
F_d (mol/yr)	3.4e9	3.4e9	3.4e9	3.4e9	3.4e9	3.4e9	3.4e9	3.4e9
$R_{87,d}$ initial ¹	0.70714	0.70714	0.70714	0.70714	0.70714	0.70714	0.70714	0.70714
F_{ht} (mol/yr)	16.4e9	16.4e9	16.4e9	16.4e9	16.4e9	16.4e9	4.1e9	4.1e9
$R_{87,ht}$	0.7030	0.7030	0.7030	0.7030	0.7030	0.7030	0.7030	0.7030
Perturbations								
Hydrothermal Flux								
LIP Initial Phase (30-31.75 Myr) ²	1.16X	1.16X	1.225X	1.225X	1.155X	1.152X	1.19X	1.185X
LIP Main Phase (31.75-35.75 Myr)	1.879X	1.861X	2.353X	2.316X	1.908X	1.884X	2.153X	2.039X
Post LIP (35.75 Myr)	1.4X	1.4X	1.6X	1.6X	1.35X	1.35X	1.4X	1.4X
Late Aptian-Early Albian rise (45.4 Myr)	1.2X	1.2X	1.3X	1.3X	1.15X	1.15X	1.2X	1.2X
Total Weathering Flux								
LIP Initial Phase (30-31.75 Myr)	1.00X	1.00X	1.053X	1.055X	1.00X	1.00X	1.04X	1.04X
LIP Main Phase (31.75-35.75 Myr)	1.25X	1.262X	1.593X	1.602X	1.30X	1.30X	1.24X	1.25X
Post LIP (35.75 Myr)	1.1X	1.1X	1.2X	1.2X	1.1X	1.1X	1.1X	1.1X
Late Aptian-Early Albian rise (45.4 Myr)	1.21X	1.21X	1.32X	1.32X	1.21X	1.21X	1.21X	1.21X

¹See Supplemental Table 3 for full list of $^{87}\text{Sr}/^{86}\text{Sr}$ values for the diagenetic flux adjusting every 2.5 Myr.

²Model times at which perturbations occur where model time 0 yr = 160.5 Ma.

Abbreviations: sw – seawater; tw – total weathering; d – diagenetic; ht – hydrothermal

Table 2. Model values for Late Jurassic-Early Cretaceous Sr cycle model where $R_{87,tw}$ is prescribed.

	Model AB	Model CD	Model EF	Model AB modern
[Sr] ($\mu\text{mol/L}$)	450	450	450	90
$R_{87,sw}$ initial	0.706849	0.706849	0.706849	0.706849
F_{tw} (mol/yr) ¹	52e9 $\rightarrow$ 63e9	52e9 $\rightarrow$ 63e9	37e9 $\rightarrow$ 48e9	52e9 $\rightarrow$ 63e9
$R_{87,tw}$ ²	Prescribed	Prescribed	Prescribed	Prescribed
F_d (mol/yr)	3.4e9	3.4e9	3.4e9	3.4e9
$R_{87,d}$ initial ³	0.70714	0.70714	0.70714	0.70714
F_{ht} (mol/yr)	16.4e9	16.4e9	16.4e9	4.1e9
$R_{87,ht}$	0.7030	0.7030	0.7030	0.7030
Perturbation Factors				
Hydrothermal Flux				
LIP Initial Phase (30-31.75 Myr)	1.16X	1.225X	1.155X	1.19X
LIP Main Phase (31.75-35.75 Myr)	1.879X	2.353X	1.908X	2.153X
Post LIP (35.75 Myr)	1.4X	1.6X	1.35X	1.4X
Late Aptian-Early Albian rise (45.4 Myr)	1.2X	1.3X	1.15X	1.2X
Total Weathering Flux				
LIP Initial Phase (30-31.75 Myr)	1.00X	1.053X	1.00X	1.04X
LIP Main Phase (31.75-35.75 Myr)	1.25X	1.593X	1.30X	1.24X
Post LIP (35.75 Myr)	1.1X	1.2X	1.1X	1.1X
Late Aptian-Early Albian rise (45.4 Myr)	1.21X	1.32X	1.21X	1.21X

¹During consistent preconditions (0-30 Myr) F_{tw} increases 1e9 mol/yr every 2.5 Myr

²See Figure 5 for single prescribed $R_{87,tw}$ values and Table 3 for list of prescribed $R_{87,tw}$ values that vary every 1-2.5 Myr. See Supplemental Table 4 for list of prescribed $R_{87,tw}$ values used to test frequency variation with Model AB.

³See Supplemental Table 3 for full list of $^{87}\text{Sr}/^{86}\text{Sr}$ values for the diagenetic flux adjusting every 2.5 Myr. Abbreviations and model time same as Table 1.

Table 3. Prescribed $R_{87,tw}$ for models AB, CD and EF for initial and reassessed (final) fluxes at the mixed 1-2.5 Myr intervals.

Age (Ma)	Model Time (Myr)	Prescribed $R_{87,tw}$ Model AB		Prescribed $R_{87,tw}$ Model CD		Prescribed $R_{87,tw}$ Model EF	
		Initial	Final	Initial	Final	Initial	Final
160.5	0	0.70816	0.70816	0.70816	0.70816	0.70871	0.70871
158	2.5	0.70827	0.70827	0.70827	0.70827	0.70883	0.70883
155.5	5	0.70845	0.70845	0.70845	0.70845	0.70898	0.70898
153	7.5	0.70857	0.70857	0.70857	0.70857	0.7091	0.7091
150.5	10	0.70856	0.70856	0.70856	0.70856	0.70911	0.70911
148	12.5	0.70858	0.70858	0.70858	0.70858	0.70907	0.70907
145.5	15	0.70859	0.70859	0.70859	0.70859	0.7091	0.7091
143	17.5	0.70864	0.70864	0.70864	0.70864	0.70915	0.70915
140.5	20	0.70870	0.70870	0.70870	0.70870	0.70919	0.70919
138	22.5	0.70871	0.70871	0.70871	0.70871	0.70914	0.70914
135.5	25	0.70880	0.70880	0.70880	0.70880	0.70924	0.70924
133	27.5	0.70886	0.70886	0.70886	0.70886	0.7093	0.7093
130.5	30	0.70890	0.70890	0.70889	0.70889	0.70933	0.70933
129.5	31	0.70894	0.70894	0.70895	0.70895	0.70938	0.70938
128.5	32	0.70907	0.70904	0.70908	0.70904	0.70954	0.70945
127.5	33	0.70900	0.70897	0.70901	0.70898	0.70946	0.70937
126.5	34	0.70895	0.70891	0.70894	0.70889	0.70936	0.70928
125.5	35	0.70891	0.70887	0.70892	0.70888	0.70936	0.70927
124.5	36	0.70863	0.70878	0.70869	0.70889	0.70897	0.70924
122	38.5	0.70856	0.70872	0.70864	0.70883	0.7089	0.70917
119.5	41	0.70848	0.70863	0.70855	0.70874	0.7088	0.70907
117	43.5	0.70839	0.70854	0.70846	0.70865	0.7087	0.70896
116	44.5	0.70847	0.70863	0.70853	0.70871	0.70878	0.70905
115	45.5	0.70857	0.70866	0.70857	0.70875	0.7089	0.70911
114	46.5	0.70869	0.70876	0.7087	0.70885	0.70907	0.70924
113	47.5	0.70870	0.70879	0.70871	0.70887	0.70908	0.70925
112	48.5	0.7087	0.70876	0.70868	0.70884	0.70904	0.70922
111	49.5	0.70869	0.70878	0.7087	0.70886	0.70907	0.70925
110	50.5	0.70864	0.70875	0.70862	0.70884	0.70895	0.7092
108	52.5	0.70857	0.70869	0.70856	0.70877	0.70887	0.70912
Weighted average		0.70862	0.70866	0.70863	0.70869	0.70906	0.70913
Standard deviation		0.000210	0.000193	0.000205	0.000200	0.000219	0.000158
Maximum difference		0.00091	0.00088	0.00092	0.00088	0.00084	0.00074
Maximum step magnitude		0.00028	0.00018	0.00023	0.00018	0.00039	0.00015
Step magnitude average $\pm$ SD (10^{-5})		7.1 ± 5.9	6.2 ± 4.0	6.9 ± 5.3	5.8 ± 4.1	8.6 ± 7.4	6.5 ± 3.8

Table 4. Final model values for Late Jurassic-Early Cretaceous Sr cycle model where $R_{87,tw}$ is prescribed and varies every 1-2.5 Myr.

	Model AB	Model CD	Model EF
[Sr] ($\mu\text{mol/L}$)	450	450	450
$R_{87,sw}$ initial	0.706849	0.706849	0.706849
F_{tw} (mol/yr)¹	52e9 $\rightarrow$ 63e9	52e9 $\rightarrow$ 63e9	37e9 $\rightarrow$ 48e9
$R_{87,tw}$²	Prescribed	Prescribed	Prescribed
F_d (mol/yr)	3.4e9	3.4e9	3.4e9
$R_{87,d}$ initial³	0.70714	0.70714	0.70714
F_{ht} (mol/yr)	16.4e9	16.4e9	16.4e9
$R_{87,ht}$	0.7030	0.7030	0.7030
Perturbation Factors			
Hydrothermal Flux			
LIP Initial Phase (30-31.75 Myr)	1.16X	1.225X	1.155X
LIP Main Phase (31.75-35.75 Myr)	1.85X	2.30X	1.90X
Post LIP (35.75 Myr)	1.55X	1.80X	1.55X
Late Aptian-Early Albian rise (45.4 Myr)	1.20X	1.40X	1.25X
Total Weathering Flux			
LIP Initial Phase (30-31.75 Myr)	1.00X	1.053X	1.00X
LIP Main Phase (31.75-35.75 Myr)	1.257X	1.593X	1.35X
Post LIP (35.75 Myr)	1.1X	1.2X	1.1X
Late Aptian-Early Albian rise (45.4 Myr)	1.15X	1.25X	1.175X
Albian Plateau (50.5 Myr)	1.10X	1.20X	1.125X

¹During consistent preconditions (0-30 Myr) F_{tw} increases 1e9 mol/yr every 2.5 Myr

²See Table 3 for prescribed $R_{87,tw}$ values.

³See Supplemental Table 3 for full list of $^{87}\text{Sr}/^{86}\text{Sr}$ values for the diagenetic flux adjusting every 2.5 Myr.

Figure 1.

Figure 2.

Figure 5.

Figure 4.

Figure 6.

Figure 7.