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Seawater Sulfur Isotope Fluctuations in the Cretaceous

Adina Paytan,[1]* Miriam Kastner,[2] Douglas Campbell,[2] Mark H. Thiemens[2]

The exogenic sulfur cycle is tightly coupled with the carbon and oxygen cycles, and therefore a central component of Earth's biogeochemistry. Here we present a high-resolution record of the sulfur isotopic composition of seawater sulfate for the Cretaceous. The general enrichment of isotopically light sulfur that prevailed during the Cretaceous may have been due to increased volcanic and hydrothermal activity. Two excursions toward isotopically lighter sulfur represent periods of lower rates of pyrite burial, implying a shift in the location of organic carbon burial to terrestrial or open-ocean settings. The concurrent changes in seawater sulfur and inorganic carbon isotopic compositions imply short-term variability in atmospheric oxygen partial pressure.

The chemical and isotopic composition of the ocean changes over time in response to fluctuations in global weathering rates and riverine loads, volcanic activity, hydrothermal exchange rates, sediment diagenesis, and sedimentation and subduction processes. All of these are ultimately controlled by tectonic and climatic changes. Accordingly, because the oceans are well mixed on time scales of thousands of years, changes in ocean chemistry reflect global changes in the relative rates of operation of the above processes. Clues about these changes can be derived from studies of the stable isotopic compositions of unaltered marine chemical precipitates, which record seawater chemistry.

Seawater contains a large amount of S ($\sim 40 \times 10^{18}$ mol) that is present, as it has been for at least the past 500 million years, predominantly as oxidized, dissolved sulfate (SO_4^{2-}) (*1*, *2*). In marine sediments, S may be present in both its oxidized state as sulfate minerals and in its reduced form as sulfide minerals. The total amount of oceanic sulfate and its isotopic composition are controlled by the input and output fluxes of S to the ocean and by their isotopic values (*1*, *2*). The largest input today is from river runoff, with $\delta^{34}S$ values between 0‰ and 10‰, depending on the relative amounts of gypsum and pyrite in the drainage basin (*3*, *4*). Volcanism and hydrothermal activity presently are small sources of S to the ocean with $\delta^{34}S$ ~3‰ (*4*). The output flux is from deposition of evaporites ($\delta^{34}S_{evaporite} \cong \delta^{34}S_{SO4}$ of seawater) and sulfides ($\delta^{34}S_{pyrite}$ on average $\cong$ –15‰, a wide range in nature) (*3*, *5*). Sulfur depleted in ^{34}S results from the large isotopic fractionation involved in bacterial sulfate reduction, the precursor for sulfide mineral formation (*5*, *6*). As a result of the formation and burial of sulfides, seawater is enriched in ^{34}S relative to any of the input sources to the ocean. The seawater $\delta^{34}S_{SO4}$ record therefore provides estimates for changes in the flux and/or isotopic composition of the input sources, as well as the relative partitioning of S between burial as oxidized and reduced reservoirs through time (*1*, *2*). Because the S sedimentary cycle is intimately related to global changes in atmospheric chemistry and climate (*7*), the seawater $\delta^{34}S_{SO4}$ record is a sensitive indicator of change in the geological, geochemical, and biological environments (*7*–*9*). In particular, the S cycle (along with C, P, and Fe) is closely linked to atmospheric oxygen levels (*2*, *7*, *10*–*12*).

A detailed $\delta^{34}S_{SO4}$ curve for the Cenozoic has been published and discussed in terms of the parameters controlling fluctuations in seawater $\delta^{34}S_{SO4}$ and the effect of these changes on global biogeochemical cycles (*13*–*17*). Here, we extend this curve to the Cretaceous, a period characterized by warm climate (*18*), high global sea levels (*19*), extensive volcanism (*20*), episodes of widespread deposition of organic carbon in black shales (*21*), and removal of inorganic carbon into chalk and carbonate platforms (*22*). Previous $\delta^{34}S_{SO4}$ data for this period are based on widely separated evaporite deposits (*23*–*25*); however, these data include large gaps and considerable scatter, and the estimated precision of the data is at best 2‰. We have generated a higher resolution (<1 million years) $\delta^{34}S_{SO4}$ curve for the Cretaceous using marine barite, a mineral that precipitates in seawater and has been shown to record the seawater $\delta^{34}S_{SO4}$ (*13*, *26*) (Fig. 1).

The seawater $\delta^{34}S_{SO4}$ in the Cretaceous, reconstructed using marine barite (fig. S1), is consistent with existing data (*23*–*25*), although it is considerably more detailed and reveals fine structure not seen in the previous isotope records (Fig. 1 and table S1). The record shows a generally lower $\delta^{34}S_{SO4}$ in the Cretaceous compared with that of the past 45 million years (*13*) (background ~19‰ versus ~22‰). Superimposed on this background are two distinct excursions toward lower $\delta^{34}S_{SO4}$. Between 125 and 120 million years ago (Ma), it falls quickly from 20‰ to 15.5‰ and remains low for ~15 million years. Starting at ~105 Ma, it increases to a maximum of ~20‰ at 95 Ma and then decreases over the next 10 million years to another minimum of 17.5‰, centered around 82 Ma. After this, minimum $\delta^{34}S_{SO4}$ returns to the background value of 19‰ (at 78 Ma) and slowly decreases during the late Cretaceous to ~18.5‰ at the K/T boundary. Both excursions, similar to the excursion in the late Paleocene (*13*), occur on a relatively short time scale (5 to 10 million years), in the range of the residence time of S in the ocean (10 to 15 million years) (*27*).

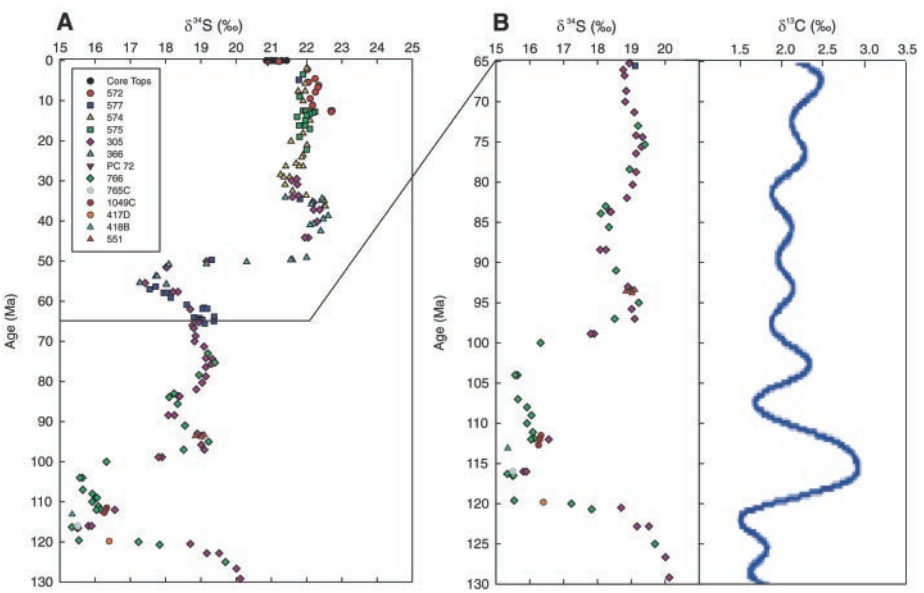


Fig. 1. (**A**) Seawater sulfate S isotope curve for the past 130 million years. (**B**) Seawater sulfate S and carbonate C isotope curves for the Cretaceous. S isotopes are from this work, and C isotopes are from (*40*).

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To explore what the $\delta^{34}S_{SO4}$ record implies about the biogeochemical S cycle, we have used a simple steady-state mass- and isotope-balance model (Eq. 1). This model represents the relation between $\delta^{34}S_{SO4}$ and the isotopic compositions and fluxes of S input to the ocean and S burial as pyrite at any given time (*9*, *17*). Using this model, we evaluate the change in each of the model parameters needed to explain the difference in background $\delta^{34}S_{SO4}$ values between the Holocene and the Cretaceous,

$$F_{py} = \frac{(\delta^{34}S_{in} - \delta^{34}S_{SO4}) \times F_{in}}{\delta^{34}S_{SO4} - \delta^{34}S_{py}} = \frac{(\delta^{34}S_{in} - \delta^{34}S_{SO4}) \times F_{in}}{\Delta^{34}S_{SO4\text{-}py}} \quad (1)$$

where F_{py} is the pyrite burial flux; F_{in} is the total S input flux to the ocean; $\delta^{34}S_{SO4}$, $\delta^{34}S_{in}$, and $\delta^{34}S_{py}$ are the S isotopic compositions of seawater, the input flux, and pyrite, respectively; and $\Delta^{34}S_{SO4\text{-}py}$ refers to the seawater-pyrite isotopic difference. Solving Eq. 1 for the Holocene with preanthropogenic values ($\delta^{34}S_{SO4}$ = 21‰, $F_{in} = 2.0 \times 10^{12}$ mol/year, $F_{py} = 0.9 \times 10^{12}$ mol/year, and $\delta^{34}S_{in}$ = 5.76‰) (*25*) yielded a seawater-pyrite isotopic difference of $\Delta^{34}S_{SO4\text{-}py}$ = 33.87‰, which lies within the range of published estimates (*25*). By inserting the background Cretaceous $\delta^{34}S_{SO4}$ (19.0‰) and letting only one of the other parameters ($\delta^{34}S_{in}$, F_{in}, F_{py}, $\Delta^{34}S_{SO4\text{-}py}$) change each time (i.e., keeping other variables at Holocene values), we calculated the values of these parameters that satisfy the model. By comparing the results with Holocene values, we obtain the maximum change needed for each parameter to explain the generally lower $\delta^{34}S_{SO4}$ in the Cretaceous.

These calculations indicate that the observed background $\delta^{34}S_{SO4}$ of 19.0‰ could be explained by (i) a lowering of the average isotopic composition of S input to the ocean ($\delta^{34}S_{in}$) from 5.76‰ to 3.76‰; (ii) an increase of 15% in the input flux of S to the ocean (F_{in} changes from 2.0×10^{12} mol/year at present to 2.3×10^{12} mol/year in the Cretaceous); or (iii) a 13% decrease in the burial rate of pyrite (to $F_{py} = 0.78 \times 10^{12}$ mol/year). The fourth parameter ($\Delta^{34}S_{SO4\text{-}py}$) value required to satisfy the model is 29.4‰, which is somewhat lower than the accepted and widely observed Phanerozoic sulfur fractionation factor (*28*). It is also considerably lower than that calculated from the average isotopic composition of Cretaceous pyrite ($\delta^{34}S_{py}$ = –21‰) (*29*), which is $\Delta^{34}S_{SO4\text{-}py}$ = 40‰. If we use the latter value in the equation, we get slightly larger changes for the other parameter ($\delta^{34}S_{in}$ = 3.36‰, $F_{in} = 2.7 \times 10^{12}$ mol/year, and $F_{py} = 0.66 \times 10^{12}$ mol/year) compared with the values calculated using the Holocene $\Delta^{34}S_{SO4\text{-}py}$.

Other lines of geological evidence place constraints on which of these four parameters are most likely to have been different in the Cretaceous (*18*, *20*, *27*, *30*, *31*). Geological evidence indicates increased ocean crust production coupled with active midplate and plate margin volcanism (*30*, *31*). Elevated levels of volcanic and hydrothermal activity will result in increased fluxes of S and a subtle shift in the $\delta^{34}S$ of S. from these sources to the ocean, as well as increased greenhouse gas input to the atmosphere. This in turn will increase both the temperatures and continental weathering rates (*20*, *27*, *30*) and is consistent with the overall warm climate and less radiogenic Sr and Os isotope records in the Cretaceous (*31*–*34*). Thus, our preferred interpretation to explain the overall lower $\delta^{34}S_{SO4}$ in the Cretaceous (19.0‰) is a shift in the total flux and isotopic composition of the S input to the ocean (F_{in} and $\delta^{34}S_{in}$). The total S flux to the ocean is composed of three major sources: weathering of continental rock, subaerial volcanic S, and marine hydrothermal activity (*25*). At present, it is estimated that these sources contribute to the ocean 1.47, 0.33, and 0.2×10^{12} mol/year, with $\delta^{34}S$ of 6.7‰, 3.0‰, and 3.5‰, respectively (*25*). A 10% increase in continental weathering flux and a 25% increase in the flux of volcanic and hydrothermal sources (to 1.62, 0.41, and 0.25×10^{12} mol/year) will change both the total flux to the ocean and the S isotopic composition of the input (2.28×10^{12} mol/year and 5.66‰, respectively). This change is sufficient to explain the lower Cretaceous $\delta^{34}S_{SO4}$. Thus, we concur that the lower Cretaceous $\delta^{34}S_{SO4}$ reflects higher volcanic and hydrothermal activities and slightly higher weathering rates (*27*). However, it must be emphasized that currently there is not enough evidence to either exclude or include the possibility that changes in the S output flux contribute to the low $\delta^{34}S$ as well.

Similar steady-state calculations for the minima $\delta^{34}S_{SO4}$ of 15.5‰ result in much larger changes (~50%) in each of the variables: $\delta^{34}S_{in}$ = 2.52‰, $F_{in} = 3.7 \times 10^{12}$ mol/year, and $F_{py} = 0.487 \times 10^{12}$ mol/year. The steady-state approximation, however, is not applicable to changes that occur on time scales similar to or shorter than an element's residence time, such as during the Cretaceous S excursions (*17*, *35*). It has been shown that steady-state approximations grossly underestimate the changes needed to explain rapid transitions (*17*). A time-dependent solution could be used (Eq. 2) in which the rate of change of $\delta^{34}S_{SO4}$ is calculated from the seawater curve. For example, we use –0.8‰ per million years, as seen between 125 and 120 Ma, and the parameters calculated above for the Cretaceous steady state to estimate the change in each variable needed to satisfy Eq. 2 (keeping all other variables constant).

$$\frac{d\delta^{34}S_{SO4}}{dt} = \frac{F_{in}(\delta^{34}S_{in} - \delta^{34}S_{SO4}) - F_{py}(\delta^{34}S_{SO4} - \delta^{34}S_{py})}{M} \quad (2)$$

Our calculations show that changes in each of the parameters (F_{in}, F_{py}, or $\delta^{34}S_{in}$), assuming seawater sulfate concentrations similar to the present, are unreasonably high. The total S flux to the ocean would have more than doubled (from 2.28 to 5.1×10^{12} mol/year), the isotopic composition of the input flux would yield unacceptably low values (–10.8‰), and the pyrite burial flux would have almost completely diminished (to 0.04×10^{12} mol/year). One possible solution that would not require huge changes in either continental weathering rates or pyrite burial is a fourfold increase in both the volcanic and hydrothermal S inputs. Such an increase would result in a total S flux to the ocean of 4.12×10^{12} mol/year (double the pre-excursion flux), with a $\delta^{34}S_{in}$ of 4.67‰.

If seawater sulfate concentrations were, however, lower than at present, as some evidence suggests (*36*), smaller but still important changes would be needed. For example, a 25% lower seawater sulfate concentration (21 mM versus 28 mM) would require either a doubling of the input flux to the ocean (to 4.5×10^{12} mol/year), or a $\delta^{34}S_{in}$ value of –7.3‰, which is very low and could only be achieved if the S in the continental weathered terrain was predominantly pyrite; or a reduction in the pyrite burial flux to less than 20% of the pre-excursion value (0.16×10^{12} mol/year instead of 0.9×10^{12} mol/year).

Each of these changes alone, even for an ocean with lower sulfate concentration, is very large and demands changes that cannot be justified by observations of current processes and their rate of operation. Accordingly, the most probable scenario is a combined change in the total S flux to the ocean, its isotopic composition, and pyrite burial. We speculate that rapid chemical weathering rates of recently erupted flood basalts and Oceanic Plateaus (*37*), in conjunction with a warmer, wetter climate, accelerated the supply of S from continental sources to the ocean. Increased volcanism and hydrothermal activity contributed to a higher S input flux and a lower S isotopic composition of this flux. In addition, the pyrite burial flux was likely lower. For example, a possible (but not unique) scenario would be a 20% increase in continental weathering rates associated with a small change in the isotopic composition of weathered rocks (from 6.68‰ to 6‰), along with a doubling of the hydrothermal and volcanic fluxes and a 55% decrease in the pyrite burial rates. These changes are large but conceivable based on the geological record and our understanding of the global biogeochemical cycle and the processes involved in this cycle, such as distribution of rocks of the continents, weathering rates, volcanism, spreading rates, and biological evolution (*1*, *7*).

The requirement to invoke a considerable change in pyrite burial has implications for the C and O cycles. Pyrite burial rates depend on sulfate, organic matter, and Fe availability. The former two are substrates used by sulfate-reducing bacte-

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ria to produce H_2S, which reacts with Fe to form pyrite (*38*). Observations suggest that in settings of "normal marine" deposition, the burial of pyrite and organic C are positively related, because organic matter is the major substrate used by sulfate-reducing bacteria (*38*). Thus, a possible explanation for the lower pyrite burial as suggested by the S isotope curve between 120 and 105 Ma could be low organic matter availability for sulfate reducers during this time interval. The geological record, however, shows that the mid-Cretaceous is characterized by ocean-wide high organic matter burial (*18*–*21*). This high organic matter burial is also supported by the higher C isotope ratios of dissolved carbonate in seawater at that time (*39*, *40*); thus, limitation of sulfate reduction by organic matter availability is unlikely to be the reason for a lower pyrite burial rate. Alternatively, pyrite formation may have been limited by iron availability despite high sulfate reduction rates (*41*), or in addition to the well-documented oceanic burial a considerable amount of organic matter burial may have taken place in continental settings where pyrite formation is limited by sulfate availability (*38*).

It is interesting to compare the S isotope curve to a compiled C isotope curve of seawater carbonates for the Cretaceous (*39*, *40*) (Fig. 1B). This comparison must be regarded only as a first-order observation because the records are not composed using the same cores, which may result in age offsets. In addition, the variability in response time of C and S as a result of the different residence times of these elements in the ocean is not considered here (*17*). It has been suggested that a general negative correlation between $\delta^{34}S_{SO4}$ and $\delta^{13}C_{CaCO3}$ exists (*42*, *43*). Indeed, the broad low $\delta^{34}S$ between 120 and 105 Ma is mirrored by high $\delta^{13}C$ between 120 and ~100 Ma. However, as previously seen for the Cenozoic, over shorter time scales (one million to several million years), the isotopic records are not negatively correlated (*13*, *44*), which suggests that the deposition of organic C and pyrite S has not been compensatory, resulting in fluctuations in atmospheric oxygen (*44*). Such changes in atmospheric oxygen have important implications for the evolution and dispersal of organisms (*16*). Alternatively, these data may imply that another process, such as changes in the cycles of Fe or P (*2*, *7*, *10*–*12*), operated to consume/produce oxygen to balance the system.

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Supporting Online Material

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Soils, Agriculture, and Society in Precontact Hawai'i

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Before European contact, Hawai'i supported large human populations in complex societies that were based on multiple pathways of intensive agriculture. We show that soils within a long-abandoned 60-square-kilometer dryland agricultural complex are substantially richer in bases and phosphorus than are those just outside it, and that this enrichment predated the establishment of intensive agriculture. Climate and soil fertility combined to constrain large dryland agricultural systems and the societies they supported to well-defined portions of just the younger islands within the Hawaiian archipelago; societies on the older islands were based on irrigated wetland agriculture. Similar processes may have influenced the dynamics of agricultural intensification across the tropics.

What determined the distribution and dynamics of intensive agriculture in tropical forest environments, before European contact? The question is controversial; some argue that the soils of most tropical forests are suited only for long-fallow shifting cultivation (*1*), whereas others contend that many rain forests have been shaped by a long history of intensive cultivation (*2*, *3*). Analyses of Polynesian agriculture are relevant to this question, because Polynesians used a variety of intensive agricultural practices in a broad range of tropical environments. Here we evaluate how climate and soil fertility defined the distribution of large rain-fed dryland systems in the Hawaiian Islands, on both local and archipelago-wide scales.

About 3000 years ago, the progenitors of the Polynesians brought a suite of crops, domestic animals, and agricultural strategies into the central Pacific, where they developed a distinctive culture that in the first millennium A.D. radiated to the margins of Eastern Polynesia (*4*). By the time of significant European contact in the late 18th century, many Polynesian economies were highly intensive, with short-fallow or irrigated agricultural systems supporting dense populations in societies with substantial social hierarchy and cultural complexity (*5*, *6*).

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