

Reference: Biogeosciences Discussion 12: 1205-1245

Authors: Markovic et al.

Title: Pleistocene sediment offloading and the global sulfur cycle

General comments:

The authors have composed a well-written manuscript describing and interpreting a high-resolution sulfur isotope record of marine sulfate determined on authigenic marine barite crystals from an Eastern Equatorial Pacific ODP site spanning the last 3 million years. The manuscript is written in a logical sequence, is well organized, easy to read and understand. Previous literature is appropriately considered and figures and tables are of good quality. Most conclusions appear well supported by the provided data and by the utilized sulfur cycling model. The analytical approaches are very sophisticated giving confidence in the presented data. The authors describe a $>1.0\%$ decrease in $\delta^{34}\text{S}$ of marine sulfate from circa 22.0% to $<21.0\%$ with the majority of the change occurring between 1.5 and 1.0 Ma. The authors use a sulfur cycle model to conclude that erosion during sea level low-stands was only partly compensated by increased sedimentation during times of sea level high-stands, an interpretation that appears well justified based on the presented data and model runs.

In my view, there are three issues in this manuscript that require some further discussion and clarification.

1) The authors suggest that the observed decrease in $\delta^{34}\text{S}$ is related to the Milankovic cycle driven change from 41 kyr (and 23 kyr) interglacial-glacial periodicity earlier in the Pleistocene to 100 kyr by 700,000 years ago. While the end of the reported $\delta^{34}\text{S}$ decrease described in this manuscript is consistent with this interpretation, the question arises why the decrease in $\delta^{34}\text{S}$ values of marine sulfate started as early as 1.5 Ma ago if it is linked to 100 kya cycles? To my best knowledge, oxygen isotope records of benthic foraminifera place this transition somewhere between 1.0 and 0.7 Ma ago (e.g. Ruddiman, W. F. (2008): *Earth's Climate*, Freeman & Co, New York), but your Figure 2 indicates that a 0.7% shift in $\delta^{34}\text{S}$ of marine sulfate had already occurred between 1.5 and 1.2 Ma ago, which appears inconsistent with the larger 100,000 yrs interglacial-glacial periodicity capable of removing much larger sediment loads from the shelf. The authors should add some explanation on the timing of this early onset of the decrease in $\delta^{34}\text{S}$ of marine sulfate reported in their paper, and possibly compare it to the $\delta^{18}\text{O}$ record of benthic foraminifera, which has reportedly been used for age-dating the samples (page 1209, line 3).

2) I am also curious why organic sulfur compounds receive so little attention in the sulfur cycling models, but assume that they play only a very minor role in the reported S fluxes. Nevertheless it should be acknowledged (e.g. on page 1217, line 10) that some sulfur is also buried in organic form.

3) And finally, in section 4.2 the authors comment on the link between pyrite oxidation, acidification-enhanced carbonate dissolution/precipitation and associated CO₂ release into the atmosphere. While I agree with the principle geochemical arguments, I am a bit puzzled about the link of the here presented data showing a $\delta^{34}\text{S}$ decrease in marine sulfate starting 1.5 mio years ago and ending 700,000 years ago, and the claim that this is partly related to CO₂ increases 600kyr and 400kyr ago. The authors should provide further arguments on the temporal connections of their S isotope record with the ice core CO₂ record in order to substantiate their argument of a causal relationship.

On a technical note, $\delta^{34}\text{S}$ should be followed by “values” ($\delta^{34}\text{S}$ values) rather than “compositions” and certainly not “ratios”.

Table A1: add units (‰) after $\delta^{34}\text{S}$ and use same number of significant digits behind the comma for reporting the results;

Figure 4, y-axis label, unit missing (‰).