Response to Reviewer #1

I have embedded specific responses to Reviewer #1's comments below. Reviewer comments are in italicized text and my responses are in plain text. Excerpts from the revised text are in bold.

-Brendan and coauthors

Anonymous Referee #1

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I made two attempts to review this paper but each time became distracted. I was successful on a third attempt -and realized that the length and detail were problematic for me.

We have shortened the paper and cut back on the level of detail overall, despite adding additional material at the request of reviewers.

An honest comment is that this reads very much like a thesis with all the style that this implies. It could very well be suitable as a monograph or for a journal that specializes in reviews, but for a mainstream science journal where space and word count are prized it is much too wordy. This is not to minimize the very extensive effort put in to this careful analysis, and there may be room for senior Editorial discretion.

I found the insights into the very strong Red Sea signals to be new and interesting (and I would assume that the Persian Gulf might be similar) and that they make great sense. I was less compelled by the extensive riverine input analysis. Those signals are there, and have been known for perhaps a century or so; but seasonal and other

We retained the Red Sea analysis while cutting back on the analysis of other riverine signals considerably. Other reviewers made similar suggestions.

temporal changes will occur on a large scale (see Figure 7) and it is likely that individual investigators will make their own adjustments for this on a local basis.

Section 4 provides a nice analysis of the influence of competing processes and I found this more useful than the regional analyses that precede it.

I think I missed any references to the very high shelf pore water alkalinity and the benthic flux results? I would guess that is more significant than the minor influence of ikaite.

Pore waters are undoubtedly important reservoirs for A_T from carbonate dissolution, but we minimize discussion of how they are distinct from other reservoirs for dissolved carbonate minerals because we have little to add to that body of research. We now refer interested readers to a review of this literature (Chen, 2002). Also, we now explicitly include pore waters in "external calcium carbonate cycling:"

External carbonate cycling refers to input of alkalinity from carbonate minerals dissolved in rivers, hydrothermal vent fluids, sediment pore waters, and submarine

groundwater discharge, and carbonate removal by biogenic carbonate burial and authigenic mineralization in sediments.

The suggestion of future work (page 18) suggests more of the same. I would have preferred to see some insights into what new experiments, field or laboratory, could be devised or hypotheses tested in some real way.

We are now provide more detail regarding our suggested future work in the conclusion. Some of which involves forward biogeochemical Earth System Models and is quite different from this study in focus, despite utilizing the Alk^* tracer.

We intend to use Alk^* for two future projects. First, Alk^* is superior to A_T for monitoring and modeling changes in marine chemistry resulting from changes in carbonate cycling with ocean acidification. $A_{\rm T}$ varies substantially in response to freshwater cycling, so Alk* trends may be able to be detected sooner and more confidently attributed to changes in calcium carbonate cycling than trends in $A_{\rm T}$. Preliminary explorations of Earth System Model output suggest time of trend emergence for the alkalinity trends discussed by Ilyina et al. (2009) could be reduced by as much as a factor of 5. Secondly, we will estimate global steady state Alk* distributions using Alk* sources and sinks from varied biogeochemical ocean circulation models alongside independent water mixing and transport estimates (e.g. Khatiwala et al., 2005; 2007). We will interpret findings in the context of two hypotheses proposed to explain evidence for calcium carbonate dissolution above the aragonite saturation horizon: (1) that organic matter remineralization creates undersaturated microenvironments that promote carbonate dissolution in portions of the water column which are chemically supersaturated in bulk, and (2) that highmagnesium calcite and other impure minerals allow chemical dissolution above the saturation horizon.