

Editor Comments to the author:

Thank you for your response to my comments. I don't think you really understood the breadth of my request so I will try to be a bit more explicit.

Your approach for alkalinity source attribution in section 2.5.2 is not robust (although I am ok with the denitrification derived estimates) as discussed in previous communication. Just to recap for clarity. Although it is likely that your alkalinity is derived from CaCO₃ dissolution, CaCO₃ dissolving is likely to have been historically deposited in the sediment and does not necessarily reflect what is currently entering as you account for in your box model. Furthermore, it seems possible that there are regions of FeS generation and TA production not accounted for in your model. Given these uncertainties, I don't think you can add the calculations you have to the box model.

I suggest you remove (or heavily edit to remove the CaCO₃ estimates) section 2.5.2 and use the discussion to argue where you believe the alkalinity is derived from. You will also need to change any text where you apportion alkalinity eg line 464.

AC: Dear Mr. Cook,

Thank you very much for your in-depth thought with regard to our manuscript. We feel we have addressed the referee comments, in particular addressing other metabolic sources of alkalinity and temporal variability in our revised version. As such, admittedly, we have problems following, in part even identifying your concerns. Please allow us to comment below:

1: Necessary and fundamental condition for any box model approach is steady-state, thus any argument of temporally varying processes/fluxes cannot be entertained by such an approach. This fundamental condition is clearly stated in equation 6. Furthermore, in our discussion we have picked up this point, amongst other sections 3.3. and conclusions.

However this fundamental condition requires that per time interval (or over the integration time of the model) inputs and outputs balance each other. As such we cannot consider dissolution of "historic" CaCO₃, as this would violate the steady-state condition.

On the other hand for our approach it is irrelevant whether CaCO₃ dissolves in the water phase or in/on the sediments, as long as we overall assume that inputs balance outputs per time step.

2: Furthermore, and as importantly, our approach pursues a two-step strategy.

First we establish dissolved mass balances and quantify a gain/loss of certain properties, such as alkalinity DIC and nitrate.

This approach relies on our observations with some clearly defined baseline conditions such as box volume, discharge etc., all of which have been clearly identified in our paper. The only, but crucially important outcome of this step are above mentioned terms of respective losses or gains – this without any attribution of responsible processes. We perpetuate this strategy in section 3.4 (methods 2.6), where we only consider above gain and loss terms, without any further source attribution.

In our second step we propose a source attribution to these processes. This is based on further observed variables, such as (directly observed) properties of POM, in detail the POC:PIC ratio, the POC:PON ratio etc. Here we show that it is possible to explain up to approx. 90% of the TA gain by dissolution of the imported CaCO₃, this with clear reference to above steady-state assumption. We explicitly state that this is to be considered as an upper bound, if we assume complete dissolution of imported CaCO₃. We also rearranged the wording in the sentence starting in line 15 to make the upper bound approach more clear.

We do not exclude and cannot exclude any other sources of alkalinity, and attribute to these source, such as denitrification, combined the remaining gain of TA, as a lower bound. Obviously these upper and lower bounds, respectively, depend on the dissolving proportion of the imported CaCO₃, again invoking steady-state conditions.

In summary, we feel that we have addressed both your and the overall positive referee comments in latest revised version "on the spot" and comprehensively. Furthermore, we also feel that we have discussed limitations, such as aspects of temporal variability or the role of other TA sources openly during the later section of the manuscript. We therefore hope that our manuscript is suitable for publication in BG now.

Yours sincerely,

On-Behalf of all co-authors,

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