

Gustafsson et al. investigated benthic TA generation under low oxygen conditions using a coupled physical-biogeochemical model BALTSEM and a reactive-transport sediment model (RTM). They discovered that Fe sulfide burial accounts for roughly a quarter of "missing" TA that cannot be accounted for using the BALTSEM model, with the rest of the missing TA potentially coming from groundwater and river input. In addition, the authors found that Fe limitation restricts the magnitude of the Fe-S burial produced TA. Using scenario modeling, this anaerobic TA source will become even smaller because of improvement of oxygen conditions and reduced Fe input from land, hence burial of reduced sulfur. As a result, the Baltic Sea will be more prone to the acidification risks.

The major concern I have is how representative the RTM model result is across the entire Baltic Sea. The authors stated that this model calculation is very expensive if multiple stations are included. It appears that they tried to extrapolate only one station (F80) to the entire Baltic basin where this is "muddy" sediments. It is known that sediments are heterogeneous and the Baltic definitely should not be an exception. Therefore, I question the validity of their data presentation and interpretation based on the RTM model.

The authors also admitted that historical TA measurement in rivers may have bias due to the lack of modern analytical standard (such as CRM) and approaches. If they can add a section of error analysis and see how that could contribute to the "missing" TA in the BALTSEM model. Given that groundwater and river could both contribute large fraction of TA budget in this area and potentially changing hydrological conditions, a lack of this analysis make the entire argument in the manuscript a bit flawed.

I would also add a table on the types of reactions involved in both the BALTSEM and RTM models. The authors discussed them in multiple places but sometime they were rambling, which makes it hard to follow.

Scientific questions:

P3 L2-L8, the background on TA budget is not clear. First Gustafsson et al. stated that there is an additional TA source of 344 Gmol/yr is needed to close the Baltic Sea TA budget, and 260 Gmol/yr cannot be explained, while 84 Gmol/yr was resolved with 18 Gmol/yr was attributed to net sulfate reduction. At this point, it seems that the problem has been resolved (net sulfate reduction). Then on L7, it seems that the authors separated FeS burial from net sulfate reduction, although FeS involves net sulfate reduction. Therefore, these budgetary terms need better explanation.

P6 L16-30, the argument is weak. If the alkalinity contribution isn't known well (L28), how would the authors feel comfortable to run the calibration and ascertain the direction of sedimentary TA flux?

P8 L23-25, First if you refer to a multiple panel figure, please try to using letters (a, b, c...) to distinguish these panels. More importantly, if sulfide is being produced, according to reaction stoichiometry the authors presented, it would lead to TA accumulation, regardless whether this sulfide goes down or not.

P9 L8-11, the statement that maximum CH<sub>4</sub>-driven sulfate reduction at 8 cm is the greatest is interesting and need better explanation. If you show sulfate profile, this may look like where the sulfate-methane interface (SMZ) is and may have nothing to do with the temporal variations of CH<sub>4</sub> production.

P9 L12 paragraph, the authors separated different depth layers to account for the reactions that have TA implications. However, they failed to show whether TA produced in these layers make it up to TA efflux to the water column. This paragraph needs better organization and clarification.

Technical comments:

Indentation of paragraphs would be useful

P2 L20-L25, these sentences are a bit confusing. "On a system scale" is mentioned twice (L20 and L24). The two examples after "For examples" are essentially the same thing, so it's not clear why the authors used "however" to connect the two sentences.

P3 L3, "unresolved" TA is 260 Gmol/yr, while in P9 L22 this number is 257.5 Gmol/yr, please be consistent.

P3 L20, the two paragraphs need a better transition. Elemental sulfur seems to be introduced into the context suddenly. Prior to doing so, the authors need to explain a bit of the type of reduced sulfur burial, i.e., sulfide (with Fe) and elemental sulfur. Relative abundance of these two types of reduced sulfur also needs to be presented based on the literature.

P4 L1, is it integrated to 25 cm "interval"?

P4 L6-L15, this should go into the Introduction, as really this should be background information, by no means materials and methods.

P4 L24, move "for that reason" to the beginning of the sentence.

P4 L25, where exactly does sigma-H<sub>2</sub>S come from?

P5 L5 paragraph, while the authors referred to a supplemental table in a published study, it would be desirable to include such information in their own supplemental materials and refer to it in the context. Otherwise the readers can easily get lost.

P5 L17, not a complete sentence.

P7 Line 12, what is "This" approach?

P7 L25 and P8 L10, why are the two sulfur burial in the same time period (1970-2009)

different?

P11 L9, where is 166 Gmol/yr in Table 5?

P11 L29 and P9 L 8, I believe 43% and 43.8% in these two places are the same thing, please be consistent, at least don't round off the number in the wrong way.

P11 Section 4.2, both unit TA (mmol/m<sup>2</sup>/yr) and overall TA flux (Gmol/yr) are presented, I'd suggest that you stick with one or both (with parentheses) to avoid confusion and don't let the readers to do the conversion.

P12 L1 and P11 L31, this sounds like freshwater environment but not typical marine since sulfate is depleted but there is still oxidized Fe.

P14 L6, not precise, it's the deposition of SO<sub>x</sub> and NO<sub>x</sub> that contribute to TA reduction, ammonia deposition first increase TA and decrease upon oxidation (or biological uptake), if no biological effect is involved, there is no TA implication, i.e., deposition followed by oxidation or uptake.

P14, the title of Section 4.5, this is not implications, rather simulations of future scenarios.

P15 L29, please update the link.

P15 L33-34, what about including possible temperature increase? It would be a better approach to simulate future pH with invariant TA, like in most studies.