Reviewer #1

I think the authors mostly addressed my comments from the first review.

The manuscript now shows the effect of the adopted new rate law more clearly (but I have one additional comment on the comparison of different rate laws and parameter tuning; please see general comments below). The other emphasized features of the model (e.g., time-dependency, 3G for OM and diffusive boundary) are novel but not necessarily new and the model's application is still limited, e.g., not ready for ocean acidification simulation. Nonetheless illustrated examples are interesting and the important aspects of early diagenetic modeling are concisely described. Given the detailed description of adopted rate laws and their tuning and because the model is written in Julia and MATLAB, the paper will be useful to the community. Overall, I think this version of manuscript could be acceptable for publication in GMD.

General comments:

Comparison of the two models with the new and traditional rate laws, however, is still arbitrary because the rate constants of both models are generally determined by tuning. One cannot tell whether the difference stated in the manuscript may disappear or not when adopting different rate constants for the two models. If the feature of the new rate law is the saturation-independent enhanced dissolution caused by step-edge retreat close to equilibrium (Section 2.2.3) and the authors want to quantify its effect, then the authors should adopt the same far-from-equilibrium rate constants for both models assuming that both are controlled by the same dissolution mechanisms at far from equilibrium. This helps suppress any artificial difference caused by different tuning by different authors and make the comparison clearer and more meaningful with respect to dissolution mechanisms. This may be done by changing the rate constants of either model, so that curves overlap one another in far-from-equilibrium region in Fig. 2.

We thank the reviewer for this positive review and for providing critical comments. We agree with the reviewer that to make the comparison more meaningful with respect to dissolution mechanisms, the rate constant of the "traditional" calcite dissolution rate law should be tuned so that it overlaps with the far-from-equilibrium dissolution rate law adopted in RADI. We have changed the dissolution rate constant for the "traditional" law from 100 % day^{-1} to 10 % day^{-1} so that far from equilibrium (at Ω_{ca} ~ 0.6, see Fig. 2), dissolution rates from both laws are equal.

We have updated the model the Model evaluation sections 3.1 and 3.2 which now show results from simulations using a "traditional" rate law with a 10 % day^{-1} rate constant. In both the north Atlantic and the southern Pacific stations, the calcite contents predicted by both rate laws are now similar. To describe this and include these updated results, we have also changed the following:

- We have updated Fig. 2, Fig. 4 and Fig. 5
- [section 2.2.3, last paragraph, added text]: The value of 10 % day-1 for the rate constant was chosen because it makes the "traditional" calcite dissolution law overlap with the RADI dissolution law, so that any differences between the two can be attributed to enhanced dissolution caused by step-edge retreat close to equilibrium.
- [section 3.1, last paragraph, added text]: When "traditional" 4.5-order calcite dissolution kinetics are implemented, calcite concentrations are similar to those predicted by RADI, but the predicted [TAlk] and [∑CO2] are slightly different, being lower (< ~10 μmol kg^{-1}) than RADI's in the top 15 cm, and higher in the deeper part of the sediment column.
- [section 3.2, last paragraph, added text]: The TAlk and ΣCO2 porewater profiles predicted by a RADI simulation using 4.5-order calcite dissolution kinetics are slightly lower (< ~40 μmol

kg^{-1}) than those using the new calcite dissolution kinetics scheme, and but the predicted calcite concentrations are slightly higher (< ~2%).

- [section 3.4, replaced last paragraph by]: In addition, we note than the choice of calcite dissolution kinetics implemented in RADI does not seem to have a large impact on TAlk and ΣCO_2 porewater profiles nor on the predicted calcite concentrations. RADI's step-edge retreat dissolution regime and its low reaction order induce calcite dissolution rates near equilibrium that are orders of magnitude higher than what is predicted in a high-order rate law (Fig. 2), but if the rate constant of a high-order rate law is tuned so that it overlaps the homogeneous dissolution rate law far from equilibrium, see Fig. 2, differences are limited. Thus, we conclude that using a 4.5-order rate law with a 10% day^{-1} rate constant or using the new, mechanistic calcite dissolution rate scheme implemented in RADI should lead to similar predictions.

Specific comments:

L26: I thought we agreed that the model is not necessarily ready for ocean acidification events.

The model is not appropriate for long term acidification events that cause large chemical erosion, but should be appropriate to accurately predict the effects of ocean acidification on the short run, e.g., on dissolved fluxes across the sediment-water interface. We have added the adjective long-duration in L.810 to articulate this. We kept L.26 as it is because events are generally considered of short duration.

L340, L385: Is it possible that different biogenic or differently dissolved/aged CaCO3 could have different saturation states at which the rate law changes (currently around 0.8), assuming that the rate law close to equilibrium is controlled by density of steps/kinks that can vary with the extent of the reaction and/or between biological species?

This is possible, but Subhas et al. (2018, Marine Chemistry) showed that the switch between both regimes occurs at a Ω_{ca} that is very similar whether foraminifera, coccoliths or inorganic calcite are being dissolved. So based on the available data, this has not been observed so far.

Sections 3.1 and 3.2: Why the new rate law predicts higher preservation of CaCO3, given the higher dissolution rate at > 0.8 saturation ratio and the bottom water saturation ratio of 0.88 and 0.85 at these sites? Or is the bottom water at these sites still far from equilibrium so that traditional rate law predicts faster rate than the updated law? To make comparison clearer I think the authors should modify one of the rate laws so that rates of both models are the same/similar at far-from-equilibrium(please also see general comments).

This is comment is not relevant anymore, as we have followed the reviewer's suggestion of changing the rate constant of the high-order law, and the updated CaCO3 concentrations are now similar.

L677-L680: These lines could be incorrect. The bottom water chemistry with the saturation ratios of 0.88, 0.85 and 0.78 indicates that the dissolution rate could be higher with the traditional rate law (Fig. 2; please also see the comment above). In order to discuss the difference between the two models with the two rate laws with excluding any artificial difference caused by tuning, the authors have to normalize the two tunable rate laws at far from equilibrium region assuming that both are controlled by the same mechanisms at far from equilibrium (please also see general comments).

We have changed the rate constant of the high-order law, and updated this part.

L686: It would be useful to the reader if the authors mention the limitation of the model to transient application here, probably with an explanation on why the authors chose such specific application examples and not e.g., ocean acidification where we expect more drastic change of sedimentary CaCO3 system. This could be mentioned earlier as well, e.g., Section 1 or 2.

We have already mentioned that in the section entitled "additional applications", we believe that this is sufficient to inform readers on that specific limitation of the model.

Reviewer #2

Concerns about modeling alkalinity and DIC (versus the individual species that make them up) have largely been addressed. I still have some concern that they are only partially correct in their assumption that this only a problem at low pH. The other issue to consider is that in anoxic coastal sediments other species (e.g., dissolved sulfide, ammonium) may also contribute to the measured alkalinity, and this can only be addressed by modeling individual chemical species and then determining alkalinity after the fact by summing the relevant chemical components. I'm not sure how to address this here since this is not a problem in this work, and only may be a problem in future work. One possible suggestion is to add a sentence like this to line 821,

"Other chemical species (e.g., dissolved sulfide, ammonium) that also contribute to the measured pore water alkalinity may also invalidate this assumption."

(or words to this affect). However, I leave that decision to the editor.

Other than that, this manuscript looks ready for publication.

David Burdige

We thank David Burdige for this comment, and we have added the suggested sentence to line 821.