General comments:

Sulpis et al. presented a new diagenesis model RADI with some examples of its application. The model adopts recently published rate laws for dissolution of calcite and allows for DBL parameterization. The model will be useful to the community especially because it is written in MATLAB and Julia. The manuscript is concisely written and easily understood. However, I have a couple of points of concern regarding the description of model and its performance evaluation and applicability.

1. Model's overall structure

The overall structure of the model is not described in detail. I could find only a few sentences, e.g., L130-132: 'equations composing RADI are based on CANDI, the method-of-lines code by Boudreau (1996b). Unlike the model of Boudreau (1996b), RADI does not solve a set of reactive-transport differential equations but instead computes the concentrations of a set of solids and solutes at each time step following a time vector set by the user.' Equations provided by the authors, however, indicate that they seem to use some time-forward finite difference method, e.g., L387: 'backward-difference discretization prevails', and thus they seem to have governing differential equations from which difference equations are derived. If so, what is the difference from CANDI with respect to the general model structure including adopted numerical method?

It is somewhat disturbing to read that the model does not solve reactive-transport differential equations (L130-132), and correspondingly the authors did not provide any governing equations. A general reactive-transport equation (e.g., Boudreau, 1996, 1997) formulates the mass conservation law dictating that mass loss/gain via transport and reactions, and mass change within each sediment layer are balanced for each species. Thus, any model (including RADI) should end up in solving reactive-transport equations although the numerical approach can vary with models. Again, however, the numerical method and its difference (if any) from those of other models (including CANDI) are not clearly described in the text.

The wording describing the model structure was indeed confusing, and we will clarify in the revised manuscript. We will replace the sentence 'Unlike the model of Boudreau (1996b), RADI does not solve a set of reactive-transport differential equations but instead computes the concentrations of a set of solids and solutes at each time step following a time vector set by the user.' by the following:

"RADI uses the same set of reactive-transport partial differential equations implemented in CANDI (Boudreau, 1996b), i.e., for each solute component,

$$\frac{\partial v}{\partial t} = \frac{1}{\varphi}\frac{\partial}{\partial z}\left(\varphi d\frac{\partial v}{\partial z} - \varphi u v\right) + \alpha(v_w - v) + \sum R$$

and for each solid component,

$$\frac{\partial v}{\partial t} = \frac{1}{\varphi_s}\frac{\partial}{\partial z}\left(\varphi_s b\frac{\partial v}{\partial z} - \varphi_s w v\right) + \sum R$$

where v is the concentration of a given component, t is time, φ is porewater porosity and φ_s is the solid volume fraction, d is the effective molecular diffusion coefficient and b is the

bioturbation coefficient, *z* is depth, *u* is the porewater burial velocity and *w* is the solid burial velocity, *α* is the irrigation coefficient, v_w is the concentration of a solute in the bottom waters and Σ*R* is the net production rate from all biogeochemical reactions for a given component. Each of these terms will be described in detail later in the Model description section.

These partial differential equations are solved numerically using the method of lines described in Boudreau (1996b). Instead of searching for steady-state solutions directly, RADI computes the concentrations of a set of solids and solutes at each depth and time steps following a time vector set by the user. The user determines the simulation time depending on the objectives, e.g., multimillennial to predict a steady state, or a few days to study the response of the sedimentary system to high frequency cyclic phenomena such as tides."

2. Model evaluation experiments

In steady-state experiments, the authors used Muds to be compared with RADI (Sections 3.1 and 3.2). However, the comparison does not make any sense to me if RADI is tuned but Muds is not, to specific sites considered in this paper.

Also, the diagenetic influence of the updated rate law for CaCO3 dissolution was discussed by comparing Muds and RADI (Section 3.4). This does not make any sense to me, either, given that ΣCO2 and TAlk production profiles from OM degradation can be different between the two models. If the authors want to discuss the effect of adopting the new rate law for calcite, they should compare two RADI simulations adopting the new and previous rate laws with individually tuned rate constants under otherwise the same boundary conditions.

We agree and will address this comment in the revised manuscript, where we will instead compare two RADI simulations for each site, one with the new $CaCO_3$ dissolution kinetics, and one with the high-order $CaCO_3$ dissolution kinetics traditionally used in models, including in Muds.

3. Model's applicability

The authors argued that RADI can be used for simulations imposing an intense ocean acidification event such as PETM (Section 4.4), but one may doubt it. Under an intense dissolution event (such as PETM), chemical erosion can happen where burial velocity can become negative at certain sediment depths. However, the burial velocity calculation scheme of RADI does not seem to allow this (Eq. (14), Section 2.3 and Fig. 1). Indeed, given that burial calculation does not seem to reflect any mass/volume changes in solid species caused by reactions, the transient simulation of RADI should be limited to the cases where the effect of solid mass/volume changes on burial rate is minor, e.g., short term experiments with minor changes in solid phase concentrations such as those in Sections 4.1-4.3. To enable the application to cases involving a significant CaCO3 dissolution, RADI has to adopt a different burial velocity calculation scheme, such as that adopted by Munhoven (2021, GMD, 14, 3603).

The reviewer is correct saying that burial velocity calculations do not reflect changes caused by reactions. In the revised manuscript, we will rewrite and state that RADI cannot deal with long-term major dissolution (erosion) events.

Specific comments:

Fig. 1. Sediment layer/point numbers within the model domain and depths assigned to sediment layers/points seem to be confused. For example, in Fig. 1 Z looks like the total number of sediment

points/layers meanwhile it is defined as the total sediment thickness in Table 1 and text. The same goes to dz.

That is true, we will update Fig. 1 to match the definitions given in the text. *Z* is the total height of the sediment column, and dz is the depth step separating each layer, both are expressed in meters.

L131-132. Quite vague description of the model. It is unclear how the model is different from or similar to CANDI (please also see general comment 1).

We agree and will update the text in the revised manuscript, see reply to comment 1.

L141. Is the threshold for dz/dt for stability of the numerical solution dependent on w?

We have tested this by changing the flux of clay (multiplying and dividing by a hundred) which in returns changes *w*, and it appears that solid burial velocity does not matter for the determination of the dz/dt stability threshold.

L354. Is 30% d-1 the most used rate constant with the reaction order of 4.5? I thought 100% d-1 is more often adopted in the literature (e.g., Archer, 1991, 1996; Archer et al., 1996).

This is correct, a rate constant of 100 % d^{-1} has been used more often in the literature. We will replace in the text and in Fig. 2.

Eqs. (11)-(14). What is the difference between x and w? Also, in Eq. (13) porewater volume fraction should be used instead of solid volume fraction. How do you calculate w∞ and u∞? I guess the authors assume w∞ = u∞ = x∞?

The equations in section contained some inaccuracies as pointed out by the Reviewer. This will be addressed in the revised manuscript. We will rewrite the text and Eqs.(11-14) as:

"The solid burial velocity at the sediment-water interface, w_0 in m a^{-1}, is given by:

$$w_0 = \left.\sum \frac{F_v \cdot M_v}{\rho_v}\middle/\varphi_{s,0}\right.$$ (...),

where F_v is the flux of a solid species at the sediment-water interface (mol m^{-2} a^{-1}), M_v is the molar mass of that solid (g mol^{-1}), and ρ_v is its solid density (g m^{-3}). The solid and porewater burial velocity at greater depth are assumed to be equal, and are computed as:

$$w_\infty = u_\infty = w_0 \, \varphi_{s,0}/\varphi_{s,\infty}$$ (...).

Thus, the porewater burial velocity, *u*, at all depths is:

$$u_z = u_\infty \, \varphi_\infty/\varphi_z$$ (...)

and the solid burial velocity, *w*:

$$w_z = w_\infty \, \varphi_{s,\infty}/\varphi_{s,z}$$ (...)."

This will not affect the model code.

Section 3.1. It does not make any sense to compare 2 models if the 2 models are not tuned to the observations in the same way (only RADI is tuned and Muds is not tuned?). If the rate constants and/or rate laws are different between the two models, I would expect different boundary conditions for 2 different tuned models.

We agree, and will instead compare two RADI simulations for each site, one with the new $CaCO_3$ dissolution kinetics, and one with the high-order $CaCO_3$ dissolution kinetics traditionally used in models, including in Muds. We hope this will address this comment.

L528-530. Is the assumption to calculate DBL thickness by Sulpis et al. (2018) consistent with RADI's assumption? In other words, isn't the calculation of DBL thickness by Sulpis et al. (2018) affected by including CaCO3 dissolution by OM-derived CO2?

In Sulpis et al. (2018) the DBL thickness is computed as a function of the bottom-water current speed and of the viscosity of seawater, and does not depend on any assumption made regarding organic matter degradation or CaCO3 dissolution. Hence this method to calculate DBL thickness is consistent with RADI's assumptions.

L549. The good reproduction of NO3 profile seems to be achieved at the cost of bad OM profile reproduction, for which Muds does seem to do a better job. I think the same goes to the O2 profile difference between the two models.

That is correct, POC concentrations are always better predicted by Muds than by RADI near the sediment surface, although it seems to be the opposite at depths.

Fig. 4. Calcite concentration does not seem to increase at the bottom, which looks weird given the oversaturation of calcite at the bottom and assuming that RADI allows for precipitation.

Calcite concentration does increase at depth but very slightly, by about 0.1 wt%. That is because, at 40 cm below the sediment surface, the predicted saturation state of porewater with respect to calcite is only at a value of ~1.05. We will precise that in the revised manuscript.

Section 3.2. The issues raised for Section 3.1 can apply to Section 3.2. L649. I guess the authors can definitely tell by repeating the same tuning experiments with RADI adopting the rate law used by Muds. (Please also see general comment 2.) Could you attribute some of differences in ⬚CO2 and TAlk profiles to the difference in OM diagenesis schemes between the models?

We agree, and will instead compare two RADI simulations for each site, one with the new $CaCO_3$ dissolution kinetics, and one with the high-order $CaCO_3$ dissolution kinetics traditionally used in models, rather than comparing with Muds directly without being able to attribute the differences to a process in particular.

Fig. 8. How can it be possible that O2 and CO2 fluctuate while OM and CaCO3 do not? Is the magnitude of fluctuation of OM and CaCO3 too small to see in the figure?

That is correct, [POC] and [Calcite] fluctuate too but the magnitude of the fluctuation is infinitely small. When, every 6 hours, the DBL is thinning for 3 hours to do the increasing tidal current speed, solutes can diffuse in or out of the sediment quicker, but solids do not have time to adjust due to the very slow accumulation rates in the deep sea.

Section 4.3. Why changes in OM degradation and CaCO3 dissolution are significant while no changes are recognized in Section 4.2? Is this only because imposed DBL changes are larger in Section 4.3? Or is the response of sedimentary system to DBL change dependent on the time rate of DBL change imposed?

On the timescale of this benthic chamber simulation (2 hours), changes in the POC degradation and $CaCO_3$ dissolution rates are actually also negligible, what is shown in Fig. 9 are TAlk (in red), ΣCO_2 (in yellow) and O_2 (in blue) fluxes. This was not labelled properly and we will add this to the figure in the

revised version. We do not impose the response of the sediment system to DBL changes, the porewater composition adjusts to DBL changes. Please note that we are currently working on a follow-up RADI study focusing on the competitive control of organic matter degradation and the DBL on $CaCO_3$ dissolution at the seafloor, in which we will discuss this in details and provide in-depth analyses.

We accept all the other suggestions and corrections, and will include them in the revised manuscript.

Brest regards,

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References

Boudreau, B.P. A method-of-lines code for carbon and nutrient diagenesis in aquatic sediments. Computers & Geosciences, 22, 479-496, 1996b

Sulpis, O., Boudreau, B.P., Mucci, A., Jenkins, C.J., Trossman, D.S., Arbic, B.K. and Key, R.M.: Current $CaCO_3$ dissolution at the seafloor caused by anthropogenic CO_2. Proceedings of the National Academy of Sciences, 115, 11700-11705, 2018