Date: July 8th, 2021

Subject: Response to Reviewer’s comments on BG-2021-4

Manuscript ID: gmd-2021-4 “OMEN-SED(-RCM) (v1.1): A pseudo reactive continuum representation of organic matter degradation dynamics for OMEN-SED”

Dear Reviewer 1,

On behalf of the co-authors, I want to thank you for the detailed and constructive review of our manuscript. In the following, we reply to each of the issues raised and explain how these will be addressed in the revised manuscript.

Sincerely yours,

Philip Pika and co-authors

General comments

The authors presented a multi-G approximation method, in which organic matter (OM) can be divided into a large number of classes with different degradation rate to represent a pseudo reactive continuum of OM, as an extension / further development of an existing analytical early diagenetic model (OMEN-SED) that is originally based on two reactive OM classes (2-G model). The approximation is based on two assumptions, namely (1) the distribution of reactivity/degradation rate of OM in marine sediments can be reasonably described by a gamma function, and (2) the vertical OM distribution in sediments is in an equilibrium status (i.e. the temporal gradient of OM is zero at any depth in sediments) so that an analytical solution of the transport-reaction equation of OM can be derived. Because the proposed multi-G approach is based on the analytical solution, which does not require solving the transport-reaction equation dynamically, therefore computational expense is not a hindering factor. This makes the proposed method appealing.

However, I have several major concerns:

Comment 1:
although the method seems to produce reasonable results, the authors did not provide convincing arguments that the proposed method outperforms the original 2-G model;

Response:

We agree with the reviewer that the manuscript would benefit from a short discussion of the advantages and disadvantages of continuum and (multi-) 2-G models. We will include this in the introduction of the revised version.

However, we would like to emphasize that the aim of the new model development is not to “outperform” the original version, but rather to offer a choice between two, equally valid OM degradation model formulations. When tuned to local conditions, both the original 2-G model and the RCM-approximation can perform equally well in reproducing comprehensive local porewater/sediment depth profiles and recycling fluxes, and both are very valuable approaches for estimating reaction rates in surface sediments (i.e. the top tens of centimeters). In addition, the 2G model approach also aligns well with the model description of the biological carbon pump (power law or (multi-)exponential equation) of most existing global biogeochemical models and earth system models.

However, for global scale applications, greater sediment depths or longer timescales, the continuum model has a number of advantages over the original 2G-Model:

- it captures the widely observed continuous decrease in OM reactivity with burial time/depth, while the 2G model converges to a constant apparent OM reactivity at depth (when the reactive OM-pool is consumed)

- it merely requires constraining two free parameters, while, multi-G models, with n fractions, generally require the specification of 2n-1 parameters (the reactivity and the relative size for the n OM classes). Thus they are generally over-parameterized and difficult to constrain on a global scale.

We will clearly state these points in the revised introduction of the manuscript and also shortly discuss the advantages and disadvantages of each approach. Furthermore, we will make reference to relevant papers that discuss this topic more extensively (e.g., Manzoni and Porporato (2009); Forney and Rothman, 2012; Arndt et al., 2013; LaRowe et al., 2020).

Comment 2:
the method is described in an unclear manner;

Response:
We will carefully revise the method section.
Please also see our responses to comments 3, 6, 7, 8 and 9 (Reviewer 1) as well as our response to comment 1 (Reviewer 2) for more details.

Comment 3:
while the method trys to fit observational vertical profiles in sediments, the boundary conditions needed for the model-data fit at some places do not reflect reality and bio-physcial laws; and

Response:
Here boundary conditions and model parameters are constrained as in Hülse et al. (2018). The only parameters that are tuned to fit the pore water profiles are the RCM parameters *a* and *v*. We will revise the text to clarify this.
Please also see our reply to comment 9 ‘specific comment on section 3.1’ (Reviewer 1) for a more detailed explanation.

Comment 4:
the precondition for the validity of the approach, namely a zero temporal gradient of OM at any depth in sediments, can hardly be met in a dynamic environment. This makes the approach of limited use for coupling to dynamic models in which sedimentation of OM is variable, which is especially true for continental margins.

Response:

This is indeed one of the main limitations of OMEN-SED(-RCM)’s analytical model approach (see section 5 in Hülse et al. 2018). OMEN-SED-(RCM) has been primarily designed for the coupling to Earth System models and to investigate long term sediment dynamics. In this context, the required assumption of steady-state is valid because the variability in boundary conditions and fluxes is generally longer than the characteristic timescales of the reaction–transport processes. However, the steady state assumption can be a limitation for the model’s applicability to shallow coastal environments.

Nevertheless, both, the previous and current version of OMEN-SED are able to reproduce observed porewater dynamics across different depositional environments ranging from the coastal to the deep ocean as evidenced by the model-data and model-model comparisons. Especially the good agreement between OMEN-SED and the fully formulated numerical RTM (BRNS, Section 3.3) shows that the steady-state assumption is not a critical limitation of OMEN-SED. In addition, as outlined in section 5: “Scope of applicability and model limitations” of Hülse et al. (2018) additional developments, such as adapting pseudo-transient dynamics will further facilitate the application of OMEN-SED to more dynamic environments.

We will re-iterate these points again in the discussion of the steady-state limitation in the revised manuscript.

Specific comments:

Comment 5:
* The method (section 2) is not described clearly.

Response:
We will thoroughly revise the model description section.
Please also see our response to comments 6, 7, 8 and 9 as well as comment 1 (Reviewer 2).

Comment 6:
(a) From eq.6, it is stated that om(k,t) represents the probability density function that determines the amount of bulk OM with a reactivity between k and k+dk at time t. As om (k,t) is a probability density function, the sum of om (k,t) across all k at any speficic t should always be 1. However, this is not satisfied in eq.7, in which om (k,0) is dependent on OM(0). Please clarify this.

Response:
Correct, om(k,t)/OM(0) is actually the probability density function. We will clarify this in the revised version.

Comment 7:
(b) I am confused by the definition and use of k. k is supposed to indicate the reactivity of OM, which is a variable. So what is the justification of eq.8 that k is determined by a, v and sediment age? I understand that the latter three parameters at any specific depth are either pre-described (e.g. v=0.15 in case studies) or derived by model-data fitting. This means that k is also fixed by these values, which is not variable any more. Further, how is the age of sediment layer at depth z derived? It seems that this quantity is another variable which needs to be solved in the method, in addition to a and v. This contradicts the statement and conclusion that only a and v need to be solved. Please clarify.

Response:
In this case, k represents the apparent OM reactivity of the bulk OM mixture. It is indeed a function of a and ν, as well as age(z) (or burial time) and thus varies with depth, z. The age(z), in turn is a function of sedimentation rate, porosity (and bioturbation intensity). We agree that this part of the model description might be confusing and will therefore carefully revise these parts.

OMEN-SED(-RCM) does not solve the RCM (and thus Eq. (4) and/or age(z)) but applies a 500G approximation of the RCM which is informed by the initial distribution of OM compounds over the reactivity spectrum as calculated by the gamma-distribution.

The bulk OM reactivity, $k_{total}(z)$, of our RCM-approximation varies with depth and can be calculated as:

$$k_{total}(z) = \frac{\sum k_i * OM_i(z)}{OM_{total}(z)}$$

In the global application $k_{total}(z)$ also varies spatially, because we apply a different set of a-v values at each grid point. We will therefore omit eq. 4 and 8 as they are not relevant for our model approach and we will clarify this point.

Comment 8:
(c) Another parameterization of k using eq. 16 clearly violates the original relationship as mentioned in (b). Please justify the validity of the method if a different parameterization of k is used.

Response:
We recognize that this part of our manuscript might be confusing and thank the reviewer for pointing this out.
This part of the manuscript describes the parametrization of Thullner et al. (2009) and Hülse et al. (2018). They apply equation 16 to derive the first-order OM degradation rate constant for their 1G model approach.
In contrast, we here use a previously published a=f(w) relationship (Equation 15) to calculate the sedimentation rate dependent apparent initial age of the OM mixture (parameter a). From the resulting initial OM distribution, we derive the discrete OM fractions and their respective first-order OM degradation rate constants for the 500G approximation. The bulk OM reactivity at the SWI of our 500G approximation (k_total in Table 2) is then calculated as:

$$k_{total}(z) = \frac{\sum k_i * OM_i(z)}{OM_{total}(z)}$$

We realize that reporting this equation in such a prominent manner leads to confusion and will thus revise this section accordingly.

Comment 9:
* In the case study 3.1, although the free variables a, v are tuned that the model produces results close to observed sediment profiles, their setting has no mechanistic connection with other environmental variables, e.g. in Table 1, it is not clear why zbio is set to 0.01 cm at depth 585m, which means that there is no bioturbation at all, but then why Dbio has a non-zero value and how these parameters are related to the setting of a and v? Also it is not clear why a has a very small value (corresponding to very small lifetime of OM, therefore quite labile component) for depth 2213 m. Compared to a very confusing setting in this case study, the setting in 3.2 (Table 2) seems more reasonable and respects reality.

Response:
For this set of simulations, all boundary conditions and parameter values are taken from Hülse et al. (2018), who adapted the boundary conditions and parameter values from the original publications where available or chose the default parameter values set in OMEN-SED, which in turn were constrained based on published values.
Hence the bioturbation coefficients for the Iberian margin sites (108, 2213 and 4298m) are taken from Epping et al. (2002). The Santa Barbara site (585m) is characterized by anoxic bottom waters (compare Reimers et al., 1990 and table 1). Therefore we set zbio to 0.01 cm (i.e. no bioturbation). Db is set to a non-zero value for mathematical reasons. In some equations underlying OMEN-SED, Db is a term in the denominator. As a consequence, we set Db to a small non-zero value to avoid dividing by zero. But, the table actually contains a typo. For the site at 585 m depth, Db is set to 1e-20 and not 0.02. We will correct this typo.

RCM parameters are constrained based on best-fit solutions to the porewater observations and are independent from the other boundary conditions listed in Table 1. The best-fit solution for the observed porewater and depth profiles at the deep site on the Iberian margin yields indeed a comparably low a value indicating an initial presence of comparably reactive components. This is in line with the comparably high (k=0.1 yrs^{-1}) first-order reaction rate constant of the more reactive pool determined by Hulse et al., 2018 and reflects the heterogeneity of OM quality on the spatial scale. Apparent OM reactivity is controlled by a complex interplay of environmental factors and OM deposited at 2000 meter depths (or more) and might be as reactive or even more reactive than OM deposited in the shallow ocean (see e.g. Fig. 15a in Arndt et al., 2013). The Iberian margin is a highly productive and dynamic depositional environment that is characterized by the development of pronounced nepheloid layers in the area Nazaré Canyon (Epping et al., 2002). These nepheloid layers extend to considerably greater water depths and efficiently transport more reactive OM laterally down the slope. Thus the intense lateral transport of OM might explain the occurrence of comparably reactive OM at greater depths.

We will carefully revise the site-specific information.

Comment 10:
* There is hardly justification for the validity of the approach in global application in section 3.3, as shown in Fig.6. In particular, the part that simulated OPD exceeds 10^3 mm is not supported by any observation.

Response:
In section 3.3, we aim to illustrate the model's ability to simulate diagenetic dynamics on the global scale. However, as pointed out in the manuscript itself, application of diagenetic models in data-poor areas, such as the global-scale, are currently limited by the lack of a general framework that would allow constraining OM degradation model parameters (e.g. see also Arndt et al., 2013).
Here, we here use the weak relationship between parameter a and sedimentation rate that emerges from a compilation of previously published a values (see Arndt et al. 2013 for detailed information). Although this relationship captures the very broad global patterns in OM reactivity, it does by no means capture the full variability of OM reactivity across different depositional environments and thus diagenetic dynamics (including oxygen dynamics and OPD).

We agree that our results show that OMEN-SED(-RCM) tends to overpredict observed OPD. This can be partly explained with:

1) the limitations inherent to the OM degradation model parametrization on the global scale
   The weak relationship between parameter *a* and sedimentation rate (w) that is applied to constrain parameter *a* for these simulations also tends to overpredict *a* (and thus underpredict OM reactivity) in shallower areas and thus also contributes to the mismatch between observations and simulation results.

2) a bias in the observational data set towards shallower OPDs.
   In fact, deep OPD of several meters to kilometers have been widely observed in the central parts of the oceans (D'Hondt et al., 2015; Murray, J. W. & Grundmanis, 1980, Roy et al., 2012). Based on observations from the South Pacific gyres, D'Hondt et al.,

> 2015 derive an empirical relationship between O_2 penetration depth and sedimentation rate as well as sediment thickness to show that oxygen may be present throughout the entire sediment sequence in 15–44% of the Pacific and 9–37% of the global sea floor. These estimates are further supported by the absence of a sulfate-methane transition zone in these areas (Eggert et al. 2018 and see Fig. 3 LaRowe et al., 2020 for global maps: https://www.sciencedirect.com/science/article/abs/pii/S0012825219305720).

We will further discuss this mismatch in the revised manuscript and will also add simulation results from an OMEN-SED-RCM run with a re-scaled version of the *a*-w relationship that accounts for the overprediction of a in shallower areas.

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