EXECUTIVE EDITOR

The main paper must give the model name and version number (or other unique identifier) in the title.

Even if we consider that our parameterization may not justify a model name and version, we agree to modify our title as :

A discrete interaction numerical model for coagulation and fragmentation of marine detritic particulate matter (Coagfrag V.1)

REVIEWER 1

Please note that the page and line numbers refer to the *Author's track-changes file* submitted alongside this *Author's response*.

Minor comments

> *Firstly, I was surprised to see links to the cloud microphysics field had not been made. These spectral bin models are used frequently e.g. Khain et al 2004 and a discussion is warranted.*

We do agree that some concepts used in our study come from aerosol physics. We recognize this when we refer to the seminal work of Gelbard et al. (1980) on aerosol coagulation, but we agree that a short discussion on more contemporary work in this field is relevant and will be added. We thank the reviewer for its suggestion.

We made the following changes in our manuscript. First in the Introduction at page 2 - line 46 :

> In atmospheric microphysic modeling where size spectral frameworks are used to represent the formation of clouds, efforts are made to use a small number of variables in 3-D simulations to prevent a drastic increase of the computational costs, to the expense of accuracy (Khain et al., 2015).

At page 3 - line 60 :

> It is indeed based on the seminal work of Gelbard et al. (1980) on the sectional representation of aerosol size distribution evolution due to collision and coagulation events, that the first coagulation models applied to marine snow emerged. Jackson and Burd (1998) extended the model of Gelbard et al. (1980) and applied it to the marine environment, pointing out the role of fragmentation to counter balance the importance of coagulation (Jackson et al., 1995; Hill, 1996).

And at page 3 - line 70 :

> Similar challenges arise in atmospheric GCMs. According to Kang et al. (2019) OGCMs with full cloud microphysics are still at an early stage in terms of understanding and simulating many observed aspects of weather and climate, and research is needed to circumvent these difficulties.

> *Line 375 states "But, the sensitivity of our model outcomes to many arbitrary constant parameters needs to be profoundly investigated". This is an extremely pertinent point and I think at least a basic sensitivity analysis should be conducted.*

We fully agree with the reviewer that a thorough sensitivity analysis must be conducted. However, the main objective of our manuscript is to present a numerical framework representing coagulation and fragmentation based on discrete interaction and to test its consistency and robustness with respect to the spectral resolution and discretization. This is specified at p. 3 lines 73-76: "*a formulation is sought that will attenuate the dependence of the results to the size discretization resolution. Numerical experiments are designed to study the dependence of the results on i) the number of size bins used to discretize a given size range (i.e. the resolution) and ii) the type of discretization (i.e. linear vs nonlinear)*". Although we are considering applications in biogeochemical modeling, what is presented remains general in scope. Carrying sensitivity analyses was also purposely left out because it is lengthy when properly done and that we currently have 31 pages already. Again, we agree that it is important and it is currently being done in a working manuscript.

Regarding the penalty function: while I appreciate the simplicity used to reconcile the errors arising from non-linearity, more tests should be carried out to confirm the applicability of the parameter choices used in the function.

The penalty function was empirically set up to demonstrate that it is required to correct for the asymmetry between the reactions. Following this idea, parameters were chosen by trial-and-error to achieve an arbitrary satisfactory reduction of the asymmetry. However, as mentioned in p. 22, lines 347-350, a more systematic inquiry of the resolution and non-linearity dependence is needed (which is ongoing and will be the subject of a subsequent paper) to find either an exact or empirical formulation of the penalty function.

The work aims to replace the simplified coagulation parameterizations currently used in OGCMs, which use several detritus compartments. While I completely agree that the approach used here is a positive step, it would be useful to demonstrate exactly why the model developed here is preferable beyond the current discussion given in the introduction. Is there evidence showing that carbon fluxes are estimated more accurately using this type of method? Can a simple experiment be carried out to show the shortfalls of the other approaches? There is going to be a computational penalty for including more tracers, so it should be shown that the sacrifice is worth it.

OGCMs that use many detritus compartments do not necessarily take into account coagulation. This requires that the detritus variables react together, which is not the case to our knowledge. Our ultimate objective is to explicitly implement the coagulation and fragmentation reactions in NPZD models coupled to OGCMs, with a number of detritus variables that is as low as possible. In some OGCM-BGC models, a depth-dependent function is prescribed to account for the degradation of organic matter as it sinks to the ocean floor and transformed into dissolved organic carbon p. 2 line 52: "*Gloege et al. (2017), e.g. exponential decay, Martin's curve, ballast hypothesis*". However, this cannot be viewed as an explicit parameterization of coagulation-fragmentation reactions. Other OGCM-BGC models consider numerous detritic variables, but they do not use the coagulation-fragmentation reaction results as a determinant of the size representation of their detritus variables. For example in Butenschon et al. (2016), detritus are only split up in size classes, related to size-specific primary or secondary producers variables, for which they are the terminal variable (where they end up when dead). In this example, once the material ends up in those detritic variables it is not able to move from one size class to another. They can however move from a particulate variable to a dissolved one. Thereafter, this matter is exported to the seafloor according to the size-dependent settling velocity. In our work, the particulate matter when placed in a size-specific detritic variable is able to move from one size bin to another following coagulation and fragmentation reactions. Once coupled to a BGC model, this will vary as a function of depth and time. To sum up, size-detritic variables will not be considered as terminal variables of the particulate matter, and matter dynamics and export rates will be fluctuating depending on the biological and physical environment. Therefore, we then think that our approach will provide new carbon export patterns in NPZD models that ultimately will be compared with the depth-dependent degradation functions of the organic matter as reference. Indeed those functions are representative of what is observed in the ocean, as they are based on field results. Although this is a really stimulating and important question, we are however unable, in the current state of our research, to carry out any experiment of comparison. This requires additional work, such as the implementation of parameters known to be variating with size (coagulation and fragmentation), and a parameterization with a physical and biological environment. This is precisely stated in the manuscript (p.24 line 385): "*Ultimately, when reliably parameterized, this model will be coupled to an upper trophic level ecological model and OGCMs that will enable addressing further questions related to the fate of particle evolution with depth.*"

Takeuchi et al. 2019 finds aggregates are bounded in size by the Kolmogorov length scale. Rather than using an arbitrary upper bound, this characteristic could be used to inform the choice of upper bound.

This is true that the Kolmogorov length scale constrains the upper bound of particle sizes and we thank the reviewer for pointing out a recent study about this. However, there are also evidences that some phytoplankton species (e.g. *Melosira Arctica*) can form 20 cm-long strands that reach the seafloor thousands of meters below the surface (Boetius et al, 2013). Despite its importance, prescribing an upper bound is beyond the scope of our paper which is to develop a numerical framework that is robust and that can be adapted to practical applications. There are multiple ways to represent the effects of turbulence in and those will be discussed in a subsequent study.

References

Boetius, A., Albrecht, S., Bakker, K., Bienhold, C., Felden, J., Fernández-Méndez, M., Hendricks, S., Katlein, C., Lalande, C., Krumpen, T., et al.: Export of algal biomass from the melting Arctic sea ice, Science, 339, 1430–1432, https://doi.org/10.1126/science.1231346, 2013.

Butenschön, M., Clark, J., Aldridge, J. N., Allen, J. I., Artioli, Y., Blackford, J., Bruggeman, J., Cazenave, P., Ciavatta, S., Kay, S., Lessin, G., van Leeuwen, S., van der Molen, J., de Mora, L., Polimene, L., Sailley, S., Stephens, N., and Torres, R.: ERSEM 15.06: a generic model for marine biogeochemistry and the ecosystem dynamics of the lower trophic levels, Geosci. Model Dev., 9, 1293–1339, https://doi.org/10.5194/gmd-9-1293-2016, 2016.

REVIEWER 2

Please note that the page and line numbers refer to the *Author's track-changes file* submitted alongside this *Author's response*.

Minor comments

> *Bin microphysical models for coagulation are widely used in cloud and aerosol physics. There are many studies that addressed this problem and some excellent reviews (Jacobson, M.Z., 2005; Khain et al., 2015). I would suggest the authors to cite and briefly discuss some papers from these research areas. This can help place the current work in a more general context, and exhibit the advances compared with studies from other fields.*

This comment echoes Reviewer 1's first comment. We do agree that some concepts used in our study come from aerosol physics. We recognize this when we refer to the seminal work of Gelbard et al. (1980) on aerosol coagulation, but we agree that a short discussion on more contemporary work in this field is relevant and will be added. We thank the reviewer for its suggestion.

The changes are similar to the ones made for Reviewer 1' first comment.

> *In Eqs. (8) and (9), the triplet operator that represents both coagulation and fragmentation for a given bin, should be discussed in more detailed. It is clear from Eq (9), that the increase in the number of particles in bin k is proportional the product of particles number concentrations in bins i and j. However, the coagulation and fragmentation rates (coagulation kernels) from Table 2 are considered constant, and not depending on particle radius, chemical composition or terminal velocity of the particles.*

Coagulation and fragmentation rates depend on particle size through a number of different processes and our model takes this into account. To better reflect this, we have modified the equations describing our model (mainly Equations 8-9) so that they use size-dependent coagulation and fragmentation rates, and we briefly summarise how these rates depend on size, citing relevant literature. It is only when we design the numerical experiments that those rates are set constant, to facilitate the interpretation of the results and the assessment of robustness to resolution and discretisation.

We made the following changes in our manuscript at page 25 - line 134 :

$$\begin{aligned}\delta C^{k}_{\boldsymbol{i},j} = \delta C^{\boldsymbol{k}}_{i,j} &= \frac{1}{2}\Big(- \mathcal{K}_{ij} n_{\boldsymbol{i}} n_j \mathcal{N}_{\boldsymbol{i}} + \mathcal{F}_k n_{\boldsymbol{k}} \mathcal{N}_{\boldsymbol{k}} \Big) \\ \delta C^{k}_{\boldsymbol{j},i} = \delta C^{\boldsymbol{k}}_{j,i} &= \frac{1}{2}\Big(- \underbrace{\mathcal{K}_{ij} n_i n_{\boldsymbol{j}} \mathcal{N}_{\boldsymbol{j}}}_{\text{Coagulation}} + \underbrace{\mathcal{F}_k n_{\boldsymbol{k}} \mathcal{N}_{\boldsymbol{k}}}_{\text{Fragmentation}} \Big).\end{aligned} \tag{9}$$

> A bold index indicates the bin on which the reaction applies (see also Fig. 2 for a visual explanation of this convention). By construction, of the total number of k particles involved in the reaction in Eq. (8), half of this number is associated with i and the other half with j (Fig. 2), which explains why we multiply all the terms in Eq. (9) by 1/2. $\mathscr{K}$ is the coagulation rate, while $\mathscr{F}$ is the fragmentation rate. Coagulation has been studied extensively in previous works both in atmospheric (Pruppacher andKlett, 2010) and oceanographic contexts (Jackson, 2001). It may be decomposed in a combination of a sticking probability and three main collision mechanisms (Kiørboe et al., 1990; Ackleh, 1997; Engel, 2000; Jackson, 2001): Brownian motion, fluid velocity shear and differential settling. Fragmentation ($\mathscr{F}$) can be driven by biology (e.g. related to zooplankton activities such as grazing (Banse, 1990; Green and Dagg, 1997), and swimming behavior Dilling and Alldredge, 2000; Stemmann et al.,2000; Goldthwait et al., 2004)) or driven by physics (e.g. scales of turbulence (Alldredge et al., 1990; Kobayashi et al., 1999)).

We, therefore, changed the coagulation and fragmentation rates initially expressed as $\mu_{\mathscr{K}}$ and $\mu_{\mathscr{F}}$ respectively in the entire manuscript by $\mathscr{K}_i j$ and $\mathscr{F} k$ when they weren't consider constant and by $\mathscr{K}$ and $\mathscr{F}$ otherwise. We added this precision at page 13 in line 211 :

> To further simplify, assume that the rate of coagulation and fragmentation are constants given by $\mathscr{K}$ and $\mathscr{F}$ respectively.

This requiring changes also in Eqs. (21) and (22) at pages 13 and 14 :

$$R_T = \sum_{p=1}^{N+1} (X_p + Y_p) = \sum_{p=1}^{N+1} Z_p$$
$$= \frac{1}{2} N (N+1) \left(-\frac{\mathcal{K}}{\mathcal{N}} \left(\frac{C_T}{N} \right)^2 + \mathcal{F} \frac{C_T}{N} \right),$$

and since $\mathcal{N} = 1$,

$$R_T = \underbrace{-\left(\frac{N+1}{N} \right) \frac{\mathcal{K}}{2} C_T^2}_{\text{Coagulation}} + \underbrace{(N+1) \frac{\mathcal{F}}{2} C_T}_{\text{Fragmentation}} \tag{21}$$

$$\delta C_{i,j}^k = \delta C_{i,j}^{\boldsymbol{k}} = \frac{1}{2} \Big(-\left(\frac{N}{N+1} \right) \mathcal{K}_{ij} n_{\boldsymbol{i}} n_j \mathcal{N}_{\boldsymbol{i}} + \left(\frac{1}{N+1} \right) \mathcal{F}_k n_{\boldsymbol{k}} \mathcal{N}_{\boldsymbol{k}} \Big)$$
$$\delta C_{\boldsymbol{j},i}^k = \delta C_{j,i}^{\boldsymbol{k}} = \frac{1}{2} \Big(\underbrace{-\left(\frac{N}{N+1} \right) \mathcal{K}_{ij} n_i n_{\boldsymbol{j}} \mathcal{N}_{\boldsymbol{j}}}_{\text{Coagulation}} + \underbrace{\left(\frac{1}{N+1} \right) \mathcal{F}_k n_{\boldsymbol{k}} \mathcal{N}_{\boldsymbol{k}}}_{\text{Fragmentation}} \Big). \tag{22}$$

We also made a correction regarding $\mathscr{K}$ and $\mathscr{F}$ units in Tables 1 and 2, pages 7 and 17 respectively.

The mechanisms for particle coagulation are not emphasized or discussed. For example, in Jackson (2001) three mechanisms for particle coagulation are discussed: Brownian diffusion, shear (laminar and turbulent), and differential sedimentation. It is not clear if coagulation and fragmentation rates are considered constant just to check the performance of the algorithm, or as some kind of approximation for different physical processes like differential sedimentation.

We agree with the reviewer that it was not clear from the manuscript whether the model was only applicable with constant rates and whether or not we recognized that there are well-known mechanisms and well-described formulations that exist. In response to the previous comment, we will revise the model formulation by adding size-dependent rates, including coagulation. Shortly after, we will recall the mechanisms driving particle collision and coagulation, namely Brownian motion, shear and differential sedimentation. It is only when we describe the numerical experiments that we will set these rates to constant values and say why we choose to do so.

Changes in the manuscript are related to the ones made for the Reviewer 2's second comment and in page 18 at line 300 :

Moreover, in order to simplify the problem and to focus only on the resolution-dependence of the framework, coagulation and fragmentation rates, $\mathscr{K}$ and $\mathscr{F}$ respectively, are set to constant values (Table 2).

In page 14, line 218, it is written that "Coagulation of particles belonging to bins 1 m and 10 m would ideally produce particle size 11 m", which is in general not true for the radius of the resulting particles after coagulation.

We are thankful that this point was raised, as we did not precise, indeed, that this rule applied when considering size as the particles' volume. We will correct this statement by referring to volume (in m3) instead of the one-dimensional size (in m).

We made the following changes in our manuscript at page 14 from lines 240 to 242 :

Consider, for example, a simple nonlinear discretization where bins are each separated by an order of magnitude (1 m^3, 10 m^3, 100 m^3, etc.). Coagulation of particles belonging to bins 1 m^3 and 10 m^3 would ideally produce

particle size 11 m^3. However, since 11 m^3 is much closer to bin 10 m^3 than bin 100 m^3 (the next larger bin) all the concentration associated with this reaction will fall into the 10 m^3 bin, thus violating volume conservation, yet conserving the concentration associated with the reaction.

In the discussion of simulation results, authors stated that "coagulation leads to a reduction of Cp in small size bins and an increase in larger ones for both LR and HR, resulting in a linearly increasing distribution of Cp over the resolved size range". And "fragmentation yields a reduction of Cp in larger size bins to the benefit of an increase in the small ones", as in general expected. However, it could be interesting to check further the performance of the model for the coagulation case only, by comparing the results obtained from the kinetic equations developed by the authors with analytical solutions of the Smoluchowski equation for a constant coagulation kernel (for example).

Good point, thank you. We made a formal comparison between Smoluchowski's approach and ours that will be presented in the revised version. In essence, there exists very few differences between the two. If we put aside the closure for the unresolved range, the only difference resides in the fact that Smoluchowski's formulation is in terms of number of particles, while ours is in terms of concentration. This slightly modifies the evolution equation since the total concentration is conserved for a given reaction in our setup while the total number of particles is not conserved in Smoluchowski's (two particles are combined to yield a single new one). Nevertheless the two formulations are equivalent and, in this context, it is difficult to imagine a setup that would allow a comparison outside of a simple bookkeeping between concentration and number of particles.

We made the following changes in our manuscript at page 12 from lines 190 to 202:

Equation (16) can be rewritten as a sum of the following series:

$$\begin{aligned}
&(\boldsymbol{p=1}): && X_1 = \delta C^2_{\mathbf{1},1} && ; \; Y_1 = \sum_{j=1}^{N} \delta C^{j+1}_{\mathbf{1},j} && ; \; Z_1 = 0 \\
&(\boldsymbol{1<p\leq N}): && X_p = \sum_{i=1}^{p} \delta C^{p+i}_{\boldsymbol{p},i} && ; \; Y_p = \sum_{j=p}^{N} \delta C^{p+j}_{\boldsymbol{p},j} && ; \; Z_p = \sum_{i=1}^{p-1} \Delta C^{\boldsymbol{p}}_{i,p-i} \\
&(\boldsymbol{p=\frac{3}{2}N}): && X_p = 0 && ; \; Y_p = 0 && ; \; Z_p = \sum_{j=N+1}^{2N} \sum_{i=j-N}^{N} \Delta C^{\boldsymbol{\frac{3}{2}N}}_{i,j-i}
\end{aligned} \tag{18}$$

where $\Delta C = 2\delta C$ when $p = 2i$ and δC otherwise. An example of a complete set of reactions with $N = 4$ and its additional bin $\frac{3}{2}N = 6$ is shown in Fig. 3.

Focusing on the resolved range only, Eqs. (16) and (18) can be combined to yield an alternative formulation of the discrete Smoluchowski equation (Smoluchowski, 1916)

$$\begin{aligned}
R_p = & \sum_{i=1}^{N} \Delta C^{p+i}_{\boldsymbol{p},i} - \sum_{i=1}^{p-1} \Delta C^{\boldsymbol{p}}_{i,p-i} \\
= & -\sum_{i=1}^{N} \frac{1}{2}(1+\delta_{ip})\left(\mathcal{K}_{ij} n_i n_p - \mathcal{F}_k n_{i+p} \mathcal{N}_p\right) + \sum_{i=1}^{p-1} \frac{1}{2}(1+\delta_{ip})\left(\mathcal{K}_{ij} n_i n_{p-i} - \mathcal{F}_k n_{i+p-1} \mathcal{N}_p\right),
\end{aligned} \tag{19}$$

where δ_{ip} is the Kronecker delta function that is equal to 1 for $i = p$ and zero otherwise. Notice that the above equation gives the rate of change of concentration, whereas the traditional formulation for the Smoluchowski equation is written in terms of number of particles. Equation 19 can thus be reformulated in terms of number of particles as:

$$\delta n_p = -\sum_{i=1}^{N} \left(\mathcal{K}_{ij} n_i n_p - \mathcal{F}_k n_{i+p}\right) + \frac{1}{2} \sum_{i=1}^{p-1} \left(\mathcal{K}_{ij} n_i n_{p-i} - \mathcal{F}_k n_{i+p-1}\right). \tag{20}$$

The factor $1/2$ in the second term ensures that the combination of two particles yields a single larger particle. This is in contrast to the concentration, for which the combination is additive (see Eq. 8).