

Final response to reviewer's comments on "Dynamic upscaling of decomposition kinetics for carbon cycling models"

We would like to thank the three reviewers for their comments. In this brief summary, we highlight the comments that in our view are most important to address should a revision of our manuscript be encouraged. In general, the reviewers commented favourably regarding the potential interest of the proposed work, but raised concerns about its applicability. We agree with this general concern and had already openly acknowledged the limitations of our approach in the original manuscript. However, we also think that a theoretical approach to link different scales in soil carbon cycling models is missing and this contribution provides a way to start bridging this gap that complements ongoing efforts by other groups.

Reviewer 1: the main concerns regard the interpretation of results (oscillations, convergence to equilibrium, sensitivity to changes in parameter values), and the establishment of a closed-form solution that can be applicable in biogeochemical models. In our response, we provide additional analyses and explanations of the results that can be included in an extended Discussion in the revised manuscript. In particular, we extended our analysis to a fourth type of decomposition kinetics used in soil C cycling models (inverse Michaelis-Menten).

Reviewer 2: the main concerns regard the validation of the proposed approach, its high level of abstraction, and the lack of representation of some physical processes known to determine heterogeneous distributions of soil substrates and microorganisms. In our response, we argue in favour of a theoretical framework, while acknowledging its limitation. A 'standard' model calibration/validation is not possible due to lack of fine-scale data, but the theoretical insights provided by our approach can still be useful. It is correct that some physical processes had not been represented, but our goal is to establish a link between macro- and micro-scale dynamics starting from an idealized system. In a revised manuscript, we would further highlight approach limitations; moreover, also in response to reviewer 3, we can include a simple representation of mass transfer as a proxy for physical transport processes that we had initially neglected.

Reviewer 3: the main concerns regard the applicability of the approach, the assumption of negligible cell-to-cell connectivity, and our interpretation of averaging and mean-field approximations. As explained in the responses above, our approach is still admittedly far from being readily applicable and we acknowledge this limitation in the manuscript. We can, however, improve the model by including mass transfer, thus addressing the second concern (new results are presented in the detailed response). Finally, we clarify our interpretations of the terms 'mean field approximation' and 'well-mixed' conditions, which might have created some ambiguities.

Detailed responses are attached below.

Response to reviewer 3

We would like to thank reviewer 3 for the review of our manuscript. Our responses are highlighted in blue font.

- 1. While the conclusions they drew are solid within their model configuration, I too share with others the concern that how this learned lesson could be translated into something universally applicable for other modelers. In particular, we in the soil biogeochemical modeling community have so far no unanimously accepted governing equation to solve like that exist for geophysical fluid dynamics, or hydrodynamics in general, where re-solving the microstructure effects can be achieved through the so called large-eddy simulation and sub-grid closure, and even field or laboratory experiments can be designed to derive parameterization schemes that are generally*

applicable for different situations. Personally, I am therefore wondering can the authors' approach become some tools that are easily accessible to others, e.g., like Markov chain Monte Carlo codes that are widely accessible through open source software?

Our methodology could be considered as a conceptual tool to understand the link of governing equations at two different scales. As mentioned by the reviewer, the lack of universally accepted C dynamics equations at different scales is one of the key issues in soil science. Use of scale transition theory is advantageous because it does not assume any predefined form of macro-scale equation, and depending upon the structure of the micro-scale model, the upscaling procedure results in macro-scale terms that define the variation across scales as a result of micro-scale properties (i.e. heterogeneities). The formulation of the micro-scale model is also uncertain, but we can test different variants of decomposition kinetics to test how the nonlinearities at the micro-scale translate into more or less nonlinear behavior at the large scales. In a revision, we would include an addition kinetic model to further expand the analysis, as described in our response to reviewer 1 and below.

The terms in the macro-scale equations that depend on micro-scale features are mathematically expressed by the second order spatial moments. Based on which second order moments appear in the upscaled equation and the kind of heterogeneity present in the system, one could start thinking about the relative importance of each term. This issue is partially covered in our answer to the comment 10 of reviewer 1 where we describe a possible strategy to obtain a second order approximation of upscaled decomposition rate. Such a second order approximation could be useful in operational models. In this contribution, however, we aim at providing a framework for upscaling that is complementary to other recent approaches, among which those referenced below (e.g., Tang and Riley, 2017).

- 2. Second I am a little bit disappointed that authors decided to ignore the interactions between different micro-grids. In physics, the successful upscaling is achieved only through the consideration of interactions. For instance, the scaling of Newton's law of momentum conservation, the derivation of center of gravity, or the scaling relation-ship between the Chapman-Enskog theory, lattice Boltzmann approach and the Navier-Stokes equation, are all hinged on the interactions between their parts. Therefore, it is not surprising at all that the authors found that their mean-field-approximation deviated significantly from their so-called full model simulations. Further, from existing scaling theories in the literature, another key of success seems to maintain the essential invariants of the system when one transits from one scale to another, yet the Michaelis-Menten kinetics they use is a crude approximation and misses some important invariant that is included in its origin law of mass action (Tang and Riley, 2017), and is deemed to show the difference they found. In addition, there's no guarantee that the mean-field equation will possess the same form as the micro-scale equation. For this, a very good example can be found in geophysical fluids, where at different scales, their governing equations are different, e.g., Gill (Atmosphere-Ocean dynamics, 1982)). Another more relevant example on decomposition is in Wang and Allison (2019).*

This comment includes two separate questions: the first is related to spatial redistribution and the second is regarding the choice of micro-scale kinetics. To address the first comment regarding the importance of spatial

interactions (or redistribution), we implemented two new versions of our model that include a redistribution mechanism – the first is based on diffusion and second is based on a generic mass transfer. The idea is to understand to what extent these spatial fluxes are important in the decomposition of soil organic carbon and their effect on the upscaling procedure we propose.

Diffusion-based three pool model

To demonstrate the effect of diffusive fluxes, we need to have an additional pool of C that is mobile, i.e. DOC. So the micro-scale model structure changes as follows,

$$\frac{dSOC}{dt} = a k_B B - V_m B \frac{SOC}{K_m + SOC}$$

$$\frac{\partial DOC}{\partial t} = (1 - a) k_B B - V_u B \frac{DOC}{K_{DOC} + DOC} + V_m B \frac{SOC}{K_m + SOC} + D_{DOC} \left(\frac{\partial^2 DOC}{\partial x^2} + \frac{\partial^2 DOC}{\partial y^2} \right)$$

$$\frac{\partial B}{\partial t} = Y V_u B \frac{DOC}{K_{DOC} + DOC} - k_B B + D_B \left(\frac{\partial^2 B}{\partial x^2} + \frac{\partial^2 B}{\partial y^2} \right)$$

$$R = \frac{\partial CO_2}{\partial t} = (1 - Y) \frac{K_{smm} DOC}{K_{DOC} + DOC} B$$

where SOC is soil organic carbon and B is microbial C (shorter notation, but conceptually the same as C_s and C_b in the manuscript). DOC is dissolved organic carbon, an additional carbon pool that is prone to transport via advection (not considered here) and diffusion. Also biomass is assumed to be transported according to a gradient-driven process with a given diffusivity. V_m and K_m are the maximum decomposition rate constant and the half saturation constant for SOC and similarly V_u and K_{DOC} are the maximum uptake rate and the half saturation constant for DOC uptake. $D_{DOC} = 2.6 E - 12 \frac{m^2}{s}$ and $D_B = 0.1 D_{DOC}$ are the DOC and biomass diffusivities. The coefficient a partitions microbial cell turnover between SOC and DOC.

Diffusion at pore scale is affected by the pore geometry and at the Darcy scale it is approximated as a function of soil moisture in unsaturated conditions. Here, we used the bulk diffusivity to simulate diffusion of DOC, and assumed that the diffusivity for biomass is 10 times smaller than that of DOC. Experiments studying the DOC pool suggest that DOC is a small fraction of soil organic matter and is quickly taken up by microbes (Schnecker et al., 2019). Therefore, it is reasonable to expect that the DOC pool will be at quasi-steady and the DOC three-pool model would be well-approximated by the two-pool model (same as in the manuscript). We obtained the parameters of the three pool model by changing the kinetics of DOC production and uptake so that the dynamics of SOC and B are comparable in both models. Figure R7 shows the time evolution of SOC, DOC and the microbial C pool in a heterogeneous system, along with the dynamics of the same variables obtained in a homogeneous system based on three-pool and two-pool models.

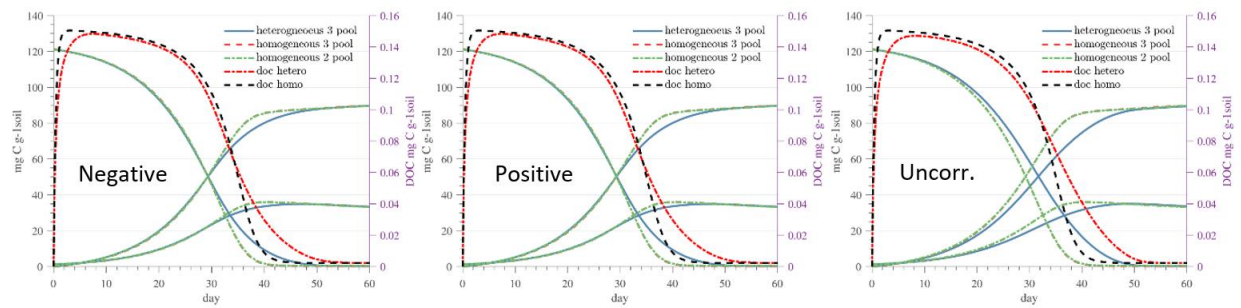


Figure R7: Temporal evolution of the mean SOC, DOC and microbial C pools when C substrates and biomass are initially negatively (left panel), positively (center) and not correlated (right). Note that the right y-axes and the black and red curves refer to DOC in all panels.

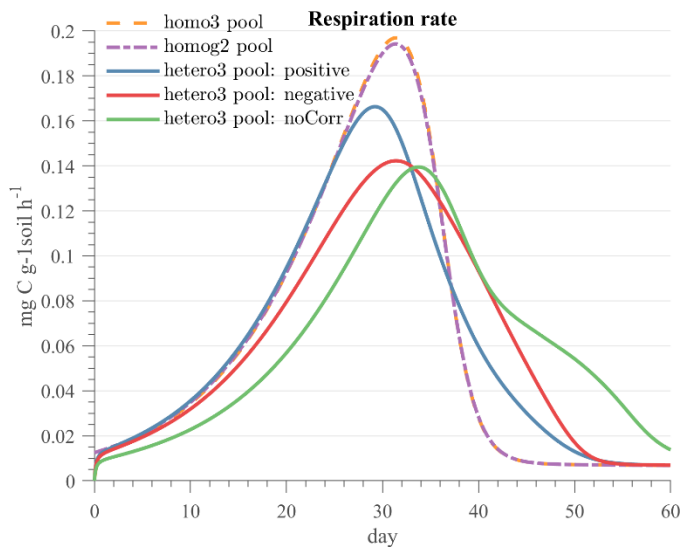


Figure R8: Mean respiration rate when C substrates and biomass are initially negatively, positively and not correlated in two- and three-pool models.

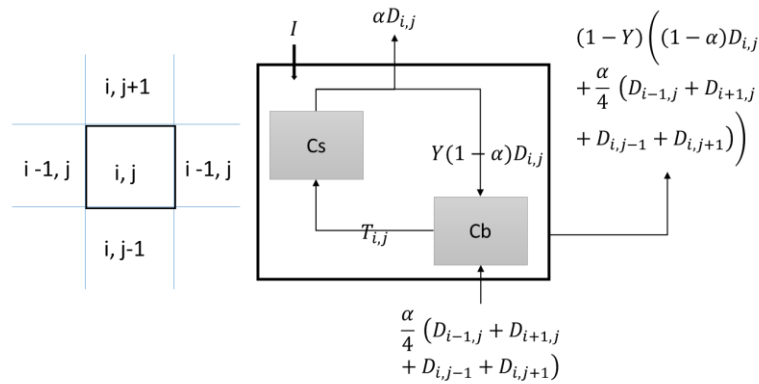
First, this analysis shows that the two- and three-pool models behave similarly when the system is homogeneous, which is expected because we constrained DOC to be turning over quickly (as suggested by empirical evidence).

Second, the three-pool model responds to the initial heterogeneous distribution of substrate and biomass showing a delayed respiration peak when the substrates are initially separated from the microbial cells (negative correlation). This result is consistent with that shown in Figure 6 of the manuscript. However, when no correlation occurs between substrates and microbes, an even longer delay emerges, in contrast to our original results in Figure 6. This suggests that diffusion does not alter the qualitative responses to micro-scale heterogeneity if microbes are in close vicinity or separated from the substrate, but could play a role in the intermediate case of no correlation. This difference between the model including diffusion and the original version without diffusion is caused by the specific initial placement of substrates and microbes. In the original

model, the initial placement did not matter, but only its statistical properties determined the dynamics (i.e., the sample size was sufficiently large). In contrast, when diffusion is included, a larger sample size becomes necessary to ensure that a specific initial configuration does not influence the dynamics. The need for a larger sample size is due to the occurrence of spatial interactions that create clusters spanning a large fraction of the domain and whose dynamics control the dynamics of the mean values of state variables and fluxes.

Mass transfer-based spatially explicit model

To explicitly include spatial fluxes across grid cells without changing the structure of the two-pool model used in the original manuscript, we implemented an alternative mass transfer mechanism. In this model, SOC is decomposed at rate D from which αD is transferred in equal amounts to the four neighboring grid cells. Hence, in each cell microbes take up C from neighboring cells at a rate $\frac{\alpha}{4} (D_{i-1,j} + D_{i+1,j} + D_{i,j-1} + D_{i,j+1})$. This choice is motivated by the observation that the products of de-polymerization are more soluble than stable organic matter and thus are more likely to be transported away from the site of decomposition. So instead of modelling DOC explicitly, we assumed that a fraction of the C flux that represents the source of soluble C is transported in other cells. This mass transfer mechanism can be interpreted as a consequence of any type of redistribution process in soils, including diffusion, dispersion or bioturbation.



If α is zero, no mass transfer occurs and the model becomes equivalent to our original two pool model. If α is greater than zero, then there is mass transfer among the grid cells. In this way, by changing the value of α , we can study the effect of spatial mass transfer flux on mean carbon dynamics. Micro-scale equations at one grid cell (control volume) take the following form,

$$\frac{dCs_{i,j}}{dt} = I - D_{i,j} + T_{i,j}$$

$$\frac{dB_{i,j}}{dt} = Y \left((1 - \alpha)D_{i,j} + \frac{\alpha}{4} (D_{i-1,j} + D_{i+1,j} + D_{i,j-1} + D_{i,j+1}) \right) - T_{i,j}$$

$$\frac{dCO2_{i,j}}{dt} = (1 - Y) \left((1 - \alpha)D_{i,j} + \frac{\alpha}{4}(D_{i-1,j} + D_{i+1,j} + D_{i,j-1} + D_{i,j+1}) \right)$$

We used this form of micro-scale model to simulate the effect of α on the averaged dynamics of decomposition for positive, negative and uncorrelated system in the same way as it was done in the original manuscript. In Fig. R9, we show the specific growth rate as a function of substrate for an uncorrelated initial distribution of substrates and microbes, and for all three kinetics– multiplicative (mult), Michaelis-Menten (MM) and inverse Michaelis-Menten (inv. MM). When $\alpha = 0$, result in Fig. R9 are same as in Fig. R6 for the uncorrelated case. When $\alpha > 0$, microbes that were initially deprived of substrate can receive it from neighboring grid cells. As a consequence of improved accessibility, given enough time microbes would consume all the substrate, whereas without mass transfer some C remains undecomposed. In other words, the persistence of substrate C we have highlighted in the manuscript in the fully heterogeneous system is lost when mass transfer provides food for microbial growth in all cells of the domain (provided enough time for transport to occur).

An interesting result emerges from Fig. R9: the inverse MM kinetics captures the effect of heterogeneity better than MM or mult, i.e. the shape of the kinetics is relatively similar to homogeneous conditions. This result might reflect some degree of scale-invariance of inverse MM kinetics.

We plan to implement this modified model in a revised manuscript, including the new figures presented here and the corresponding discussion.

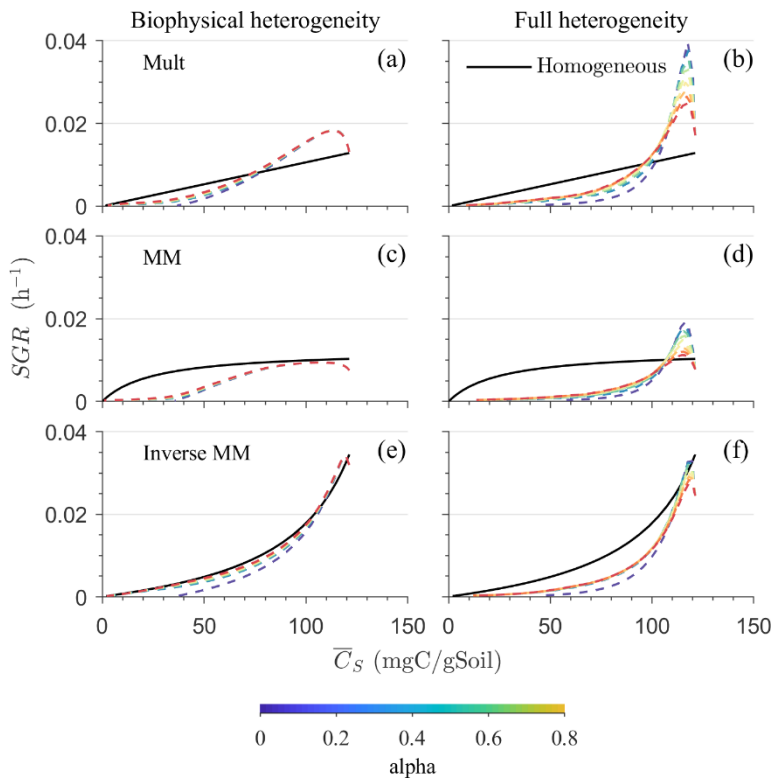


Figure R9: Effect of biophysical (left column) and full (right column) heterogeneity on the mean specific growth rate (SGR) as a function of mean substrate C (\bar{C}_S) for an uncorrelated initial distribution of substrates and microbes. The tree horizontal panels are for (a,b) multiplicative, (c,d) Michaelis-Menten and (e,f) Inverse Michaelis-Menten kinetics. Different colors represent varying values of α . Time progresses from right to left, as substrate C is depleted.

3. *Third, I feel the authors have some misunderstanding about the mean-field theory and the meaning of well-mixed soil condition. In fact, the scaling problem we are facing here is very similar like the situation hydrologists encountered in upscaling the soil moisture and soil matric potential relationship in the 1970s-1980s. Using statistical theory, they were able to derive closed analytical relationships (e.g., Mualem, 1976) to inform important soil water retention curve formulations to be derived from empirical data (e.g., van Genuchten, 1980). Therefore, whenever moisture-pressure relationships are included in soil biogeochemical models, some microstructure is included in the so-called mean-field-theory based model (although I should admit that the authors did not consider soil moisture in this study). **Or put this straightforwardly, mean field theory does not rule out the inclusion of microstructure, as was demonstrated in the recent up-scaling study of substrate affinity parameter (Tang and Riley, 2019), and the study of turbulence (e.g., Takahashi, 2017).** In the same vein, a well-mixed soil can also have microstructure, and be properly parameterized. In fact, the latter is what motivated the dual-porosity or the multiple-Rates Mass Transfer models, which have enjoyed many successful applications (e.g., Haggerty and Gorelick, 1995).*

We do not argue against macro-scale models that account for micro-scale heterogeneities in a lumped way – in fact, we hope that scale transition theory can help moving in that direction (complementing other approaches as in the cited paper by Tang and Riley). However, admittedly we are not quite to the point of proposing a closure approach that would provide closed-form equations such as the cited water retention curves.

By “mean field approximation”, we indicate the C fluxes calculated with the mean values of the state variables (e.g., as in Melbourne and Chesson, 2006), and not a mean-field theory that accounts for micro-scale processes. By “well-mixed” conditions, we refer to conditions where diffusion is faster than reaction, providing spatially uniform concentrations. This is different from assuming that e.g., random pores are ‘well-mixed’ in a soil – an assumption used to derive the cited water retention curves. We can clarify the use of these terms in a revised version to avoid any confusion.

In Tang and Riley papers, heterogeneity is introduced by the reaction network adopted for the decomposition of substrate; in other words one or more substrates can be decomposed via one or more enzymes. The question of interest in that case was: what would be the form of the decomposition function in order to account for the reaction network complexities? In this sense, we agree that a well-mixed system can have different types of substrates and enzymes, and one could apply the method developed by tang and Riley to calculate the overall decomposition function. The same philosophy was adopted by Michaelis and Menten to derive their approximated enzymatic reaction equation.

However, this question is different from what we ask here. Our contribution investigates what would happen to the decomposition function if substrate and microbes (or enzymes) are physically co-located or isolated. Spatial heterogeneity is the subject of investigation rather than the heterogeneity of the reaction network in a multi-substrate and multi enzyme system. For our purposes, the assumption of Michaelis-Menten kinetics serves the purpose. In case of chemical heterogeneity where we actually define different substrate qualities by having spatially varying kinetics parameters, the substrate decomposition in a grid cell (control volume) is performed according to the kinetic parameters associated with that cell. In other words, there is no transport of microbes (or enzymes) across grid cells that would create a multi-species system. If there was transport of chemically-different compounds, kinetic laws accounting for complex reaction networks should be used (e.g., SUPECA kinetics in Tang and Riley, 2017). Even for the spatially explicit model based on mass transfer presented earlier, simple forward or inverse MM kinetics would be enough because only a fraction of decomposition flux is transferred across the grid cells and we can assume that the compounds being transported are similar, not requiring more sophisticated kinetic laws.

While we did not implement SUPECA or other complex kinetics (note that we do not model enzymes explicitly), in a revision we can include inverse Michaelis-Menten kinetics in addition to the other kinetics. This would allow exploring the behavior of another type of micro-scale decomposition model. We are also motivated to analyze MM and inv. MM kinetics because they are commonly used and in our view modelers tend to underestimate possible scaling issues with these nonlinear functions.

Preliminary results obtained with the inv. MM kinetics are shown above.

References

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