

Revised manuscript “Dynamic upscaling of decomposition kinetics for carbon cycling models”

We have previously uploaded the point-by-point response to the reviewer comments as author comments AC1-4. This document lists the modifications made in the revised manuscript to reflect reviewers’ comments and our responses. Our arguments in support to the responses and additional analyses that are not included in the revised manuscript (but that are relevant to address the comments) are found in the author comments AC1-4.

Part of mathematical text in the marked-up manuscript did not render well because of latexdiff, for clarity of these texts please refer to the revised manuscript. Page and line number references correspond to the revised manuscript.

Reviewer 1 comments (Thomas Wutzler):

We would like to thank Thomas Wutzler for an encouraging and detailed review of our manuscript. In the following, we list the changes (highlighted in blue normal font) made in the revised manuscript in response to Dr. Wutzler (denoted by italic font).

General comments

1. *Methodology description: the way of providing spatial moments to analytic equations did not become clear to me (P17L11). I assume, you computed the quantities for sufficiently close time points from the distributed model, and provided a smoothing function depending on time as input to the solver for the analytic equation system.*

Please see our response in the interactive discussion.

2. *An overview of the approach would be helpful: 1) Express each equation of state variable change of each individual location based on the spatial mean of the pool sizes and the deviations from it at local scale. And 2) Apply a spatial averaging over the obtained equations, resulting in an equation composed of terms of the mean pool sizes and the spatial covariance of the pools and heterogeneously distributed parameters.*

The schematic Figure 1 has been improved to provide a better graphical overview of the approach, and at the beginning of the theory section the rationale was outlined in clearer terms as suggested (see, P5 L18-23):

“To obtain the macro-scale dynamics we employ two approaches: (i) a numerical approach based on grid-scale simulations followed by spatial averaging (upper panel Fig. 1) and (ii) an analytical approach based on scale transition theory (lower panel Fig. 1). The first, computationally demanding approach requires solving the micro-scale equations at each cell of the domain grid. Spatial averages and variances are thus calculated numerically over the domain at each time point in the simulation. With the analytical approach, the dynamic equations are first averaged and then solved directly for the mean state variables. The obtained analytical expressions are used to interpret the results of the numerical simulations.”

3. *I tried to check the math, but did not always come to the same results (see detailed comments, eq. A7)*

This was a typing mistake. To address this comment, the expression has been corrected in Table 2 (P14) and Eq. A7 (P36).

The corrected expression is $-\frac{2 k_{s,mm} K_M \bar{C}_B}{(K_M + \bar{C}_S)^3}$.

In the discussion I would like to read about several points:

4. *Slowdown of decomposition: To my opinion the slowdown of decomposition despite plenty of available substrate (Fig 7d) is a very important feature/insight of the model. A very simple model (albeit still required input of time series of heterogeneity variances) can explain why we can find very old potentially quickly decomposable SOM. The reasons should be explained in more detail (right skewed distribution of decomposition rate, low probability of co-occurrence of high substrate concentration, ...)*

To address this comment, a paragraph at the end of section 4.1: ‘Predicted effects of spatial heterogeneity on decomposition’ has been added at P29L16-22,

“Our analysis suggests that the persistence of SOM in heterogeneous systems may be a consequence of the micro-scale heterogeneity in soil carbon cycling. In the transient simulations with biophysical heterogeneity, persistence is a result of spatial disconnection between substrate and microorganism, captured in our framework by a low probability of co-location at the beginning of the simulation. In the transient simulations for the fully heterogeneous systems, persistence is a result of the combined effects of low probability of co-location and high probability of low decomposition rate constant at the beginning of the simulation. The heterogeneity in substrate quality thus explains the higher persistence of SOM in the fully heterogeneous system compared to the biophysically heterogeneous system.”

5. *Oscillations at multi-annual time scale: Observations of such a phenomenon are very rare. I once argued that we do not see such modelled oscillations with microbial explicit models because of superposition of dynamics across many pores. Here, such spatial heterogeneity is the cause of fluctuations.*

Please see our response in the interactive discussion. We now highlight the occurrence of these fluctuations in Section A4 (see response below).

6. *Development of the heterogeneity / damping of oscillations (Fig 4): The systems develops to a steady state without any more oscillations. Is the initial heterogeneity developing in direction of homogeneity? Probably not because the simulated SOM stocks differ from the homogeneous system. What is the spatial distribution and covariance between substrate, biomass, and quality after 60 years? Is there a covariance pattern that is stable? I suggest putting another two panels to Fig. 2 showing microbial and substrate distribution at year 60.*

To address this comment, the following text has been added at P19L14-16,

‘In systems including both biophysical and full heterogeneity, the sums of the HOT are stable in the long term, once the steady state has been reached. This was confirmed by running the model for 100 years.’

7. *Role of disturbances: What happens if you simulate a disturbance (homogenization) after the system is near steady state? Does this start the oscillating pattern again?*

This comment was previously misinterpreted. If disturbances are re-introduced after ‘new’ steady state has been reached in a previously heterogeneous system, then yes, oscillations would reappear because this new

steady state would be spatially homogeneous (recall that C input is homogeneous on the domain). Any perturbation near this ‘new’ steady state would cause fluctuations to reappear.

To address this comment, the following text has been added at P19L17,

‘Furthermore, any additional perturbation of the new steady state caused by an external factor will re-introduce the fluctuations.’

8. *Magnitude of the heterogeneity effects: In Figure 4, the effects look large, because the axis ranges from 5 to 7, but aside from the initial disturbance, the effect is only about 1/10 of the steady state. Are there reasonable parameter combinations where the effect is larger? Or do we not need to care this much about heterogeneity at steady state?*

To address this comment, new analyses have been performed. The respective results have been added to the manuscript as a part of the appendix – Appendix A4: ‘Sensitivity of fluctuations to changes in k_M in scenario 1’, at P40.

‘We performed two sensitivity analyses in which we altered the kinetic constant parameter for the multiplicative decomposition model k_M : 1) decreasing k_M in the biophysical heterogeneity–positively correlated C_s and C_b (Fig. A1) and increasing the heterogeneity of k_M (by increasing its standard deviation) in the full heterogeneity case (Fig. A2). From Fig. A1, it is clear that decreasing the rate constant increases the amplitude and wavelength of the oscillations. As shown in Fig. A2, increasing the heterogeneity of the rate constant increases the amount of undecomposed substrate C compared to a lower degree of heterogeneity (Fig. 4). This pattern can be explained using the analytical expression of the steady state substrate C (see Eq.(A16) in Appendix A3). For the increased heterogeneity case shown in Fig. 4, we used values of a and b as listed in TableA3 for biochemical heterogeneity 1 and multiplicative kinetics, where a and b have the same meaning as in Eq. (A16). The analytical expression for the steady state, evaluated with these values of a and b, results in exactly the same steady state of substrate C as simulated by the distributed model (i.e., 15mgC/gSoil).

These fluctuations are similar to those noted in earlier papers using spatially lumped models (Manzoni and Porporato, 2007; Sierra and Muller, 2015). These papers showed that the occurrence and amplitude of the fluctuations depend on the kinetic parameter values, as is the case here.’

9. *2D system: Are the insights transferable to a 3D system. What would you expect to change? Since, there is currently no transport and interaction between the cells, I infer that aside from maybe slightly different development of the initial correlations, the dynamics should stay the same. The macro-scale equations are not affected, as I understood.*

Please see our response in the interactive discussion.

10. *More complex systems: The analytical scale transition approach worked nicely with the basic simple model. With more complex models that include many more heterogeneous parameters it will be difficult to impossible to close the model with all the combination covariances (the factorial grows very fast). Can you*

describe a strategy to determine which combinations are important and which combinations can be neglected? When have we sufficiently including more and more heterogeneities?

To address this comment, the following text has been added at P33L9-12,

‘Along similar lines, how many terms in the Taylor expansion should be retained at each level of this hierarchy remains an open question. It is also possible that the dynamics at the micro scale in combination with C redistribution lead to low values of higher order moments, thus allowing us to neglect higher order terms-- because substrate consumption, mortality of the microorganisms, and transport contribute to smoothing spatial gradients.’

Admittedly, we do not have a clear answer to this comment.

11. Time scale: I am especially interested in modelling decadal to longer-term SOM dynamics. Are the multi-annual oscillations important for the longer term dynamics? Do you expect heterogeneity to change with global change in the longer term? What is the advantage of describing the changed steady state with heterogeneity (Fig 7d) with heterogeneity inputs compared to effective model parameters? I see some advantages, but it would be nice to clarify them in the paper.

Please see our response in the interactive discussion. Despite the interest in these discussion points, we opted for keeping the revised Discussion streamlined and did not cover as initially suggested these topics for the sake of space (the manuscript is already long and discussions along the lines suggested would be rather speculative with the type of model we are using).

Specific comments:

12. eq. 4 ..6: Your simple basic model refers to the Schimel and Weintraub 2003, who actually used and suggested an inverse MM kinetics $D = k_s C_s C_b / (kM + C_b)$. It would be nice to amend your work by this decomposition equation.

To address this comment, new analyses based on inverse MM kinetics were performed. Results and modification to existing figures are listed as following:

- Figures 1 and 3 now include inverse MM kinetics
- Table 2 now lists the macroscopic rate of decomposition for inverse MM kinetics in all three heterogeneous cases
- An additional figure, Fig. 7 has been added to represent the time evolution of state variables in scenario 2
- Figure 8 has been modified by adding two new panels for inverse MM kinetics
- The respective results are now discussed at several locations – in particular: P21, L13-29.
- Derivation of mean rate of decomposition for inverse MM kinetics has now been presented in appendix on P38, L5

- An additional figure, Fig. A6 has been added to represent the time evolution of SOTs for biophysical heterogeneity in scenario 2

13. *P11L11: The sentence does not make sense to me. The variance itself is not always negative. Probably you meant: “This term is always negative because the variance of the spatial substrate distribution is a positive quantity and ...”*

To address this comment, the text has been modified as suggested at P12L6,

‘The spatial variance term is always negative because the variance of the spatial substrate distribution is a positive quantity and the partial derivatives multiplying the variances are negative in all decomposition functions that saturate at high substrate concentration.’

14. *P12L15ff: May state that therefore the mean field approximation is exact and spatial variance of this parameter has no effect on the macro-scale dynamics.*

To address this comment, the text has been modified as suggested at P13L7-9,

‘Similar derivations can be done for the microbial mortality rate ($F = T$). The Taylor expansion of microbial mortality is simpler because we assume T to follow first order kinetics implying that all the second order terms are equal to zero. Therefore, the mean field approximation is exact and spatial variance of this parameter has no effect on the macro-scale dynamics’

15. *P12L19: This paragraph comes a bit surprising without context. Why do you look at SGR?*

To address this comment, the text has been modified at P13L11,

‘To illustrate how macro-scale decomposition kinetics are affected by spatial heterogeneity, we define a macro-scale specific growth rate (SGR), which is calculated by dividing the mean respiration rate by mean microbial C in the system’

16. *P14L11ff: Potential for moving to appendix. Only the information starting from P15L5 is important*

To address this comment, a part of section 2.4: Initial 2D random fields of SOM and kinetic parameters has been moved to appendix— Appendix A2 at P38, L6. Please see the revised manuscript for clarity.

The starting paragraph of section 2.4 Initial 2D random fields of SOM and kinetic parameters will be changed as follows:

‘Two-dimensional spatially heterogeneous distributions of substrates and microbial C were generated to run the distributed model. The obtained distributions were based on following constraints: i) the total amount of organic C is set, ii) the total amount of microbial C is 1% of total organic C , iii) the maximum amount of C in a cell is set (Eq. (A12)), and iv) some grid cells have no microbial biomass. For details to the field generation procedure, see Appendix A2.’

17. P14L19: What is (fg)? I could not find the explanation. It is used several times in the text eq. 26, 27 and appendix figure and table captions.

Please see our response in the interactive discussion.

18. P16L10ff: I suggest to give more meaningful names to the scenarios instead of numbers. E.g “Steady simulation” and “High Substrate Simulation” (also update Fig 3).

The names of the scenarios have been changed as suggested. Now they read as, Steady state simulation (SS) and High substrate simulation (HS). Figure 3 and the text throughout the manuscript have been modified to reflect the change.

Response to reviewer 2 (Ali Ebrahimi):

We would like to thank Ali Ebrahimi for the review of our manuscript. Our responses are highlighted in blue (normal font). Some of the comments point to the approach limitations – indeed, this is a theoretical study the findings of which are hard to validate because there is no data at a fine-enough resolution. We would like to emphasize that the value of the proposed approach is to provide a framework for studying heterogeneity effects on measurable C fluxes and stimulate discussion in this area.

Major concerns:

1. *It is unclear to what extent the parameterization proposed in the analytical kinetic model could be experimentally validated. My major problem is that some of the quantities do not have real biogeochemical or physical meanings in which could be experimentally measured. For instance there is an emphasis on the second order moments as state variables used to close the model; however it is hard to think how such variable could be experimentally measured.*

In addition to our response in the interactive discussion, this comment has been addressed by adding the following text at P30L19-26.

‘Further, an experimental validation of the present work should stem from designing a microscale experiments using artificial porous media with different degrees of heterogeneity. Recent application of microfluidics in soil science (Stanley et al., 2016; Aleklett et al., 2018) could allow isolating the effect of spatial heterogeneity. If any difference is observed among heterogeneous systems, then our framework could be used to attribute these differences to spatial heterogeneity at the micro-scale. While the proposed mathematical framework is conceptually useful, it is thus challenging to test. Nevertheless, the prediction that co-location of microorganisms and substrates promotes decomposition is consistent with and explains theoretically the results of recent experiments (Don et al., 2013; Schnecker et al., 2019).’

2. *The type of model and scenarios proposed in this study are relevant and could potentially address some of the inconsistency in our field measurements but it could only be possible if the model could establish a systematic link to relevant abiotic and biotic factors observed in the field. While in the discussion authors have tried to relate some of the scenarios in the study to soil aggregation or pore connectivity and an entire subsection is dedicated for that, I still find that the **modeling framework is too abstract that makes the explanations quite speculative** and hard to think to what extent the decomposition rate may vary under realistic settings.*

Please see our response in the interactive discussion.

3. *The model could potentially describe some of the underlying abiotic and physio-logical mechanisms that shape the decomposition dynamics but in the current form of the manuscript this has not been explored. For instance, I was wondering to what extend half saturation to substrate and decomposition rate constant (K_M and K_s) are shaping the dynamics observed in the model. I would guess if lower K_M or high K_s would have been chosen the heterogeneous scenarios would have converged faster to the homogenous one.*

In addition to our response in the interactive discussion, to address this comment (and in combination to our response to Reviewer 1), we have included a new sensitivity analyses on the kinetic rate constant in Appendix A4. The added text is reported in our response to Reviewer 1 above; new figures A1 and A2 have also been included to illustrate the findings.

Minor concerns:

4. *I was wondering to what extend the fluctuating environmental condition (for instance fluctuating in carbon distributions) could play a role in shaping the carbon decomposition dynamics. Do you expect to see faster convergence to homogenous scenario in high intensity fluctuations?*

Please see our response in the interactive discussion.

5. *The system size that is modeled in grid based network is rather small. The number of grids, or pores equal to 10000, is basically enough to model an aggregate with the size of 0.5mm. I was wondering if this size is sufficiently large to capture heterogeneities in the soil? **For instance inter aggregate pores or macropores?***

Please see our response in the interactive discussion.

6. *Following up on the results for negative correlations, **I was wondering how much physical inaccessibility of the carbon to microbes could be relevant for the soil systems?** For instance most of the carbon protections in soil are often driven by soil aggregation and creation of anoxic microsites. In a broader term, the counter gradients created by carbon and other necessary substrates for carbon degradation could lead to inaccessibility of the carbon for microbes and not necessary physical inaccessibility. This is a phenomenon that has been previously shown in soil aggregates that due to creation of anoxic zones, the carbon configuration does not play a role in carbon consumption (Ebrahimi and Or, 2018 GCB) and in other studies showing carbon protection by aggregation (e.g., Keiluweit et al., 2017 Nat. comm.).*

Please see our response in the interactive discussion. Moreover, we added the following short paragraph in the Discussion to address this comment (see, P32 L18-21):

“Including C redistribution as a simple mass transfer process does not allow studying how soil structure affects macro-scale dynamics by creating and maintaining heterogeneous distributions of resources and oxygen, such as in soil aggregates (Keiluweit et al., 2017; Ebrahimi and Or, 2018). These patterns result from the interaction of transport and reaction processes that the proposed idealized models cannot capture.”

Response to reviewer 3

We would like to thank reviewer 3 for the review of our manuscript. In the following, we list the changes (highlighted in blue normal font) made in the revised manuscript in response to Reviewer 3 (denoted by italic 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?*

[Please see our response in the interactive discussion.](#)

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).*

[To address this comment, new analyses based on mass-transfer model have been performed. New results and modifications to existing figures are listed below:](#)

- [The micro-scale model in Fig. 1 has been modified to account for C redistribution via a simplified mass-transfer approach](#)
- [Equations 2-4 have been added to represent a generic mass-transfer model on P8, L2-4](#)
- [An additional figure, Fig. 9, has been s added to represent the effect of C redistribution among grid cells](#)
- [The new results are discussed on P23, L9-14](#)

- Discussion points based on the new mass-transfer model have been added at several locations, specifically on P29, L11

‘Increasing local connectivity among grid cells moderately reduces the effect of spatial heterogeneity on the macro-scale variables and fluxes’

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).*

In addition to our response in the interactive discussion this comment has been addressed by new results based on inverse MM kinetics, as also mentioned above. This compromise allows maintaining analytical tractability, while avoiding the inclusion of enzyme dynamics in the model, which would alter significantly the model structure. Moreover, we now refer in several points to the work by Tang and Riley (2013, 2017), which is indeed a good example of upscaling, though with a focus on reaction network rather than space per se. For example, the following paragraph has been added to the Discussion (see, P31 L9-12) :

“Conceptually, this approach is similar to upscaling chemical reaction networks to obtain a compact kinetic law that only depends on the concentrations of reactants and products (Tang and Riley, 2013, 2017). However, here we focus on spatial heterogeneity rather than on the complexity of chemical reactions. In a more complete upscaling approach, both sources of micro-scale variability should be taken into account.”

Dynamic upscaling of decomposition kinetics for carbon cycling models

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Abstract. The distribution of organic substrates and microorganism in soils is spatially heterogeneous at the micro-scale. Most soil carbon cycling models do not account for this micro-scale heterogeneity, which may affect predictions of carbon (C) fluxes and stocks. In this study, we hypothesize that the mean respiration rate \bar{R} at the soil-core scale (i) is affected by the micro-scale spatial heterogeneity of substrate and ~~microbes~~ microorganisms and (ii) depends upon the degree of this heterogeneity. To

5 assess theoretically the effect of spatial heterogeneities on \bar{R} , we contrast highly heterogeneous conditions with isolated patches of substrate and ~~microbes~~ microorganisms versus spatially homogeneous conditions equivalent to those assumed in most soil C models. Moreover, we distinguish between biophysical heterogeneity, defined as the non-uniform spatial distribution of substrate and ~~microbes~~ microorganisms, and full heterogeneity, defined as the non-uniform spatial distribution of substrate quality (or accessibility) in addition to biophysical heterogeneity.

10 ~~Three~~ Four commonly used formulations for decomposition kinetics (linear, multiplicative~~and~~, Michaelis-Menten, and inverse Michaelis-Menten) are considered in a coupled substrate-microbial biomass model valid at the micro-scale. We start with a 2D domain characterized by a heterogeneous substrate distribution and numerically simulate organic matter dynamics at each cell in the domain. To interpret the mean behavior of this spatially-explicit system, we propose an analytical scale transition approach in which micro-scale heterogeneities affect \bar{R} through the second order spatial moments (spatial variances

15 and covariances).

It was not possible to capture the mean behavior of the heterogeneous system when the model assumed spatial homogeneity, because the second order moments cause the heterogeneous system to deviate from the behavior attained under homogeneous conditions. Consequently, \bar{R} in the heterogeneous system can be higher or lower than the respiration of the homogeneous

system, depending on the sign of the second order spatial moments. This effect of the spatial heterogeneities appears in the upscaled nonlinear decomposition formulations, whereas the upscaled linear decomposition model deviates from homogeneous conditions only when substrate quality is heterogeneous. Thus, this study highlights the inadequacy of applying at the macro-scale the same decomposition formulations valid at the micro-scale, and proposes a scale transition approach as a way forward to capture micro-scale dynamics in core-scale models.

1 Introduction

Soil organic substrates and microorganisms are heterogeneously distributed in the soil medium (Nunan et al., 2002; Peth et al., 2014; Raynaud and Nunan, 2014; Rawlins et al., 2016). The importance of this heterogeneous distribution in soil organic matter (SOM) dynamics has been shown both experimentally and in modeling studies. Early experimental results show that the mineralization of SOM is affected by the non-uniform distribution of the substrates within macro and micro pores (Killham et al., 1993). The recognition that spatial location of substrates and microorganisms constrains decomposition and thus C persistence is causing a paradigm shift from the previous emphasis on chemical composition of organic substrates to a focus on the biophysical environment in which decomposition occurs (Schmidt et al., 2011; Don et al., 2013; Schnecker et al., 2019). Soil pore structure is emerging as a fundamental property that integrates these biophysical constraints on decomposition (Dun-
gait et al., 2012; Falconer et al., 2015; Fraser et al., 2016). The biophysical and biochemical properties of the pore structure such as pore connectivity, tortuosity of water and air diffusion pathways, and adsorption/desorption, limit the ~~accessibility of organic substrates to decomposers~~ access decomposers have to organic substrate. As a result, these micro-scale constraints create a spatially heterogeneous landscape with highly variable distributions of substrate and microbial C. In the following, we refer to this type of variability as micro-scale heterogeneity.

Despite the importance of micro-scale heterogeneities, most SOM decomposition models are based on reaction kinetics that are valid for reactions in well-mixed media, including C cycling schemes implemented in ecosystem and Earth system models. In well-mixed systems, the mean concentrations of substrate and microbial C, and the rates defined using these mean values are assumed to be representative of the system. Most existing SOM models embrace this assumption regardless of whether they are microbial implicit (i.e., based on first order kinetics) or microbial explicit (i.e., based on multiplicative and enzyme kinetics) (Manzoni and Porporato, 2009). This approach is often referred to as mean-field approximation and is meant to describe spatially averaged SOM dynamics at soil core- to plot-scales. There is an underlying, but untested, assumption that the kinetics that are valid under well mixed conditions at fine scales also hold at larger scales, where conditions are often far from well-mixed. For this assumption to hold, a spatially averaged C flux should be equal to the average flux when organic C is

uniformly distributed throughout the system. This is not the case when C concentrations are heterogeneously distributed and the kinetics are nonlinear (Chesson, 1998; Melbourne and Chesson, 2006; Morozov and Poggiale, 2012; Van Oijen et al., 2017). For example, even in the simple case of only two soil patches, the overall C fluxes follow more complex behaviors than within an individual homogeneous patch, requiring the use of kinetics that differ from those applied at the micro-scale (Manzoni et al., 2008). The use of the same decomposition kinetics across a wide range of spatial scales is therefore questionable in systems that are spatially heterogeneous and regulated by nonlinear kinetics.

To understand at which scale a model developed for well-mixed conditions is expected to work, both the spatial scale at which heterogeneities become important and the scale at which homogeneity can be assumed must be identified. The average inter-cell distance in soil is in the order of 10 μm (Raynaud and Nunan, 2014) and the median length of spatial correlation of SOM varies between approximately 40 and 175 μm (Rawlins et al., 2016). Furthermore, it has been argued that the pore class with diameters between 30 to and 150 μm ~~has been argued to be~~ is the most important for microbial activity (Kravchenko and Guber, 2017). This heterogeneity occurring at scales from ~ 10 to 200 μm , is generally neglected in C cycling models. Below the ~ 50 μm scale, diffusion time scales can be assumed to be faster than advection and reaction time scales (Watt et al., 2006). Thus, it can be argued that below ~ 50 μm the assumption of homogeneity is likely to hold, while it is no longer valid above this threshold (see Section 2.1.1 for details). If homogeneity cannot be assumed, how should decomposition kinetics be described in soil cores or at larger scales that include strong spatial heterogeneity?

Including micro-scale heterogeneities in the kinetics of SOM models is recognized as a much needed advancement in the field (Manzoni and Porporato, 2009; Sierra and Muller, 2015; Wieder et al., 2015), though only a few attempts have been made in this direction (Ebrahimi and Or, 2016; Van Oijen et al., 2017). In contrast, there are several examples of upscaling schemes for chemical reactions networks (Tang and Riley, 2013, 2017). The challenge is therefore to develop spatially up-scaled models that describe SOM decomposition at the macro-scale while taking into account the micro-scale heterogeneities. Mathematically, this upscaling problem is equivalent to spatial averaging of the mass balance equations based on the well-mixed assumption written at the micro-scale.

Three types of upscaling approaches are often used for dynamical systems such as those used to describe soil biogeochemical processes: (i) spatial averaging of known numerically simulated C flux fields, (ii) definition of effective parameters to capture fine-scale heterogeneity, and (iii) scale transition theory or volume averaging of the equations at the micro-scale. Spatial averaging of simulated dynamics at the micro-scale is common (Allison, 2012; Kaiser et al., 2014; Yan et al., 2016) (Allison, 2012; Kaiser et al., 2014; Yan et al., 2016; Wang and Allison, 2019), but this approach does not lend itself to analytical solutions that would offer insights into the effects of heterogeneity on macroscopic properties. The effective parameter

approach, more common in sub-surface hydrology (e.g. Dagan (1987)), has been used to relate the macroscopic decomposition rate to the characteristic parameters of micro-scale heterogeneity, but only in a minimal ‘lumped’ model (Manzoni et al., 2008). The estimated effective parameters tend to be specific to studied scenarios and difficult to generalize. Here, we focus mainly on the third method based on scale transition theory, because this approach provides a dynamic link between micro- and macro-scale using spatial moment approximations (SMA). Using scale transition theory, it is possible to obtain an analytical, but approximate representation of dynamics at the macro-scale by accounting for the nonlinear dynamics at micro-scale.

Scale transition theory is based on spatial averaging of the dynamical equations themselves (as opposed to averaging known fluxes as in point (i)). ~~This~~ For example, this approach has been used to study predator-prey population dynamics at the patch and regional scales (Bergström et al., 2006; Englund and Leonardsson, 2008; Barraquand and Murrell, 2013). The macroscopic (regional) population dynamics is controlled by the mean population densities of predator and prey, which in turn relate to the spatial statistics of population densities at the micro-scale (patch). Similar approaches are also used in hydrology to calculate average hydrologic fluxes when soil and micro-climatic conditions are spatially heterogeneous (Albertson and Montaldo, 2003; Fatichi et al., 2015), and in groundwater hydrology to derive transport equations at the Darcy- or field-scale (in this field the approach is called ‘volume averaging’ (Dentz et al., 2011)). We are aware of only one study using similar techniques to scale up C and N fluxes in ~~soils~~ from plot to regional scale (Van Oijen et al., 2017). Specifically, an empirical nonlinear function was used to link methane and nitrous oxide fluxes to soil moisture and temperature ~~at~~ in each grid cell (corresponding to the micro-scale model) and the scale transition theory was applied to calculate the mean fluxes at the regional scale. However, an explicit expression linking fluxes to C pools at any time point is not always available. In most C cycle models, the fluxes are calculated by solving first the mass balance equations for the C pools (i.e., a system of differential equations). Therefore, to proceed, these differential equations at micro-scale must be scaled-up. This upscaling exercise is expected to yield a set of differential equations describing the mass balances of the spatially averaged C compartments, including kinetics for the macro-scale C fluxes that depend on the degree of micro-scale heterogeneity.

Using scale transition theory, here we develop a general theoretical approach to link micro- and macro-scales in SOM decomposition models. ~~With this approach, we demonstrate the effect of heterogeneity and nonlinearity at the micro-scale on macroscopic decomposition rates.~~ Two types of micro-scale heterogeneity are identified and accounted for: biophysical and biochemical. Biophysical heterogeneity is caused by the non-uniform spatial distribution of substrate and ~~microbes~~ microorganisms (i.e., heterogeneous distribution of the state variables), and biochemical heterogeneity is a result of spatial variations in substrate quality and thus turnover rates (i.e., heterogeneous distribution of the values of kinetic constants). With the proposed upscaling approach, we test the hypotheses that the rate of decomposition (i) is affected by the micro-scale spatial

heterogeneity of substrate and microbial C and (ii) depends upon the degree of spatial heterogeneity. Scale transition theory is applied to ~~three~~four types of micro-scale decomposition kinetics commonly employed in C cycling models: conventional linear, multiplicative ~~-, and (M)~~, Michaelis-Menten (MM), and inverse Michaelis-Menten (IMM). Considering these ~~three~~ kinetic laws allow us to assess the consequences of neglecting spatial heterogeneities in the most common C cycling models.

5 Our specific objectives are

1. To develop an analytical upscaling solution for a two pool C model
2. To quantify the impact of different spatial structures of substrate C_s , microbial biomass C_b and kinetic parameters k on the C dynamics
3. To compare the results of a spatially-explicit heterogeneous model with the homogeneous equivalent as a function of the
10 degree of heterogeneity

While the proposed upscaling approach is general, we apply it in this contribution to scale up pore-scale processes to the scale of a small soil core or laboratory soil sample. These theoretical developments can be applied to SOM models employed to study respiration and microbial responses to perturbations at this relatively small spatial scale, or in models describing dynamics at a larger scale over relatively uniform spatial domains.

15 2 Methods

2.1 Theory

We distinguish between ‘micro-scale’ equations valid at the small scale where the well-mixed assumption holds, from ‘macro-scale’ equations valid at a larger scale of interest, which result from spatial averaging of the microscopic equations. While our derivations are general, in the presented model setup and results, we interpret ‘macro-scale’ as the scale of a small soil core.

20 The goal of spatial upscaling is to derive the macro-scale soil C dynamics by spatial averaging of the micro-scale dynamics.

~~We~~

To obtain the macro-scale dynamics we employ two approaches: (i) a numerical approach based on grid-scale simulations followed by spatial averaging (upper panel Fig. 1) and (ii) an analytical approach based on scale transition theory (lower panel Fig. 1). The first, computationally demanding approach requires solving the micro-scale equations at each cell of the

25 domain grid. ~~The mean behavior is then estimated by first order spatial moment approximation, which corresponds to the spatial averaging of the micro-scale state variables C_s and C_b (and associated C fluxes)~~ Spatial averages and variances are thus

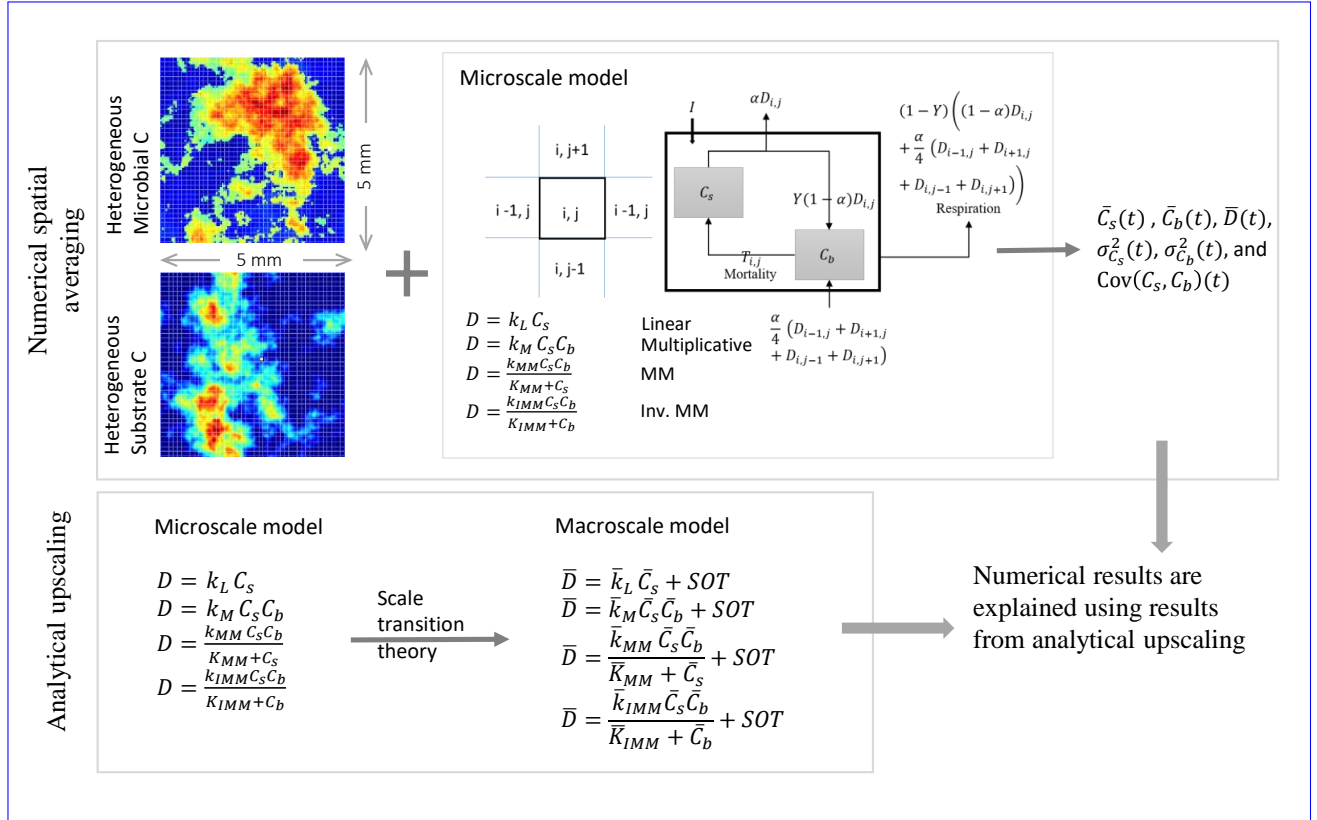


Figure 1. Schematic of the two upscaling approaches used to study the C dynamics at the macro-scale. Numerical spatial averaging (top panel): the micro-scale model is applied at each grid cell of the heterogeneous domain; and the mean C pools (substrate and microbial biomass), their mean fluxes, and second order spatial moments ($\sigma_{C_s}^2, \sigma_{C_b}^2, \text{Cov}(C_s, C_b)$) are estimated by Eq. (24)–(29) at each time step. This approach is referred to as ‘distributed model’. Analytical upscaling (bottom panel): the micro-scale model decomposition flux is dynamically scaled up using scale transition theory, which provides the mean C fluxes as a function of mean C concentrations (mean-field approximation) and second order spatial moments representing the degree of heterogeneity. The deviations from the mean-field approximation are denoted as ‘second order terms’ (SOT) in the expressions for the mean decomposition fluxes (\bar{D}), where overbar represents mean quantities). The numerical results obtained from the distributed model are explained using the mathematical expression derived from analytical upscaling. This upscaling scheme is applied to three-four types of decomposition kinetics (linear, multiplicative, Michaelis-Menten, and inverse Michaelis-Menten), shown at the bottom of the lower panel.

calculated numerically over the domain at each time step. This numerical approach is equivalent to running a distributed model where the biophysical laws describing the system are known at the micro-scale and applied throughout the entire domain. We define point in the simulation. With the analytical approach, the dynamic equations are first averaged and then solved directly for the mean state variables. The obtained analytical expressions are used to interpret the results of the numerical simulations.

5 To proceed, the spatial average operator for our 2D domain as is defined as,

$$\overline{\chi(t)} = \frac{\int \int \chi(x, y, t) dx dy}{\int \int dx dy} \approx \frac{1}{N_x N_y} \sum_{i=1}^{N_x} \sum_{j=1}^{N_y} \chi_{i,j}(t), \quad (1)$$

where the double integral extends to the whole 2D domain, χ is a generic variable (C_s or C_b) or C flux, and N_x and N_y are the number of grid cells in the x and y direction. The second equality allows estimating χ using the simulated time series of variable of interest in each grid cell (denoted by $\chi_{i,j}$). In contrast to numerically solving the problem at each grid cell, the
10 second approach derives the dynamics of the macro-scale variables and fluxes using scale transition theory, discussed in the following sections.

2.1.1 Micro-scale model of soil carbon dynamics

The dynamics of soil organic C in a homogeneous medium are characterized by specific reaction kinetics that define organic C fluxes, and the number and arrangement of the soil C pools. For simplicity, we use a two pool model that subdivides organic
15 carbon into two pools: (i) soil organic carbon substrate (C_s) and (ii) microbial biomass carbon (C_b) (Manzoni and Porporato, 2007; German et al., 2012). This simple structure was selected because it is at the core of most microbial explicit models (Zelenev et al., 2000; Schimel and Weintraub, 2003). The typical time scale of diffusive fluxes is given by $\tau_{diff} = x^2/D$ where x is the length scale of space discretization and D is the diffusion coefficient (Hunt and Manzoni, 2015) and the typical time scale of reactive fluxes is given by the turnover time of the substrate; i.e., τ_{react} . The ratio of the two time scales defines
20 the Damköhler number, $Da = \tau_{diff}/\tau_{react}$, which provides the relative importance of mass transport of substrate via diffusion vs. reaction (Dentz et al., 2011). For a relevant substrate such as glucose, D is the order of $10^{-11} \text{ m}^2/\text{s}$ (Watt et al., 2006), the turnover time is in the order ~ 1 day and the length scale of the order of $\sim 50 \text{ } \mu\text{m}$. With these values, $Da \ll 1$, which characterizes a reaction-limited system in well-mixed conditions. The result of this approximated calculation would not change with reaction time scales in the order of a few hours. Thus, the well-mixed assumption is valid at the scale of a pore $\sim 50 \text{ } \mu\text{m}$
25 and we refer to this model as a ‘micro-scale model’ (Fig. 1). The general mathematical description of the microscale model is given by-

To explicitly include spatial fluxes across grid cells, we implemented a generic mass transfer mechanism. This mechanism is implemented by assuming that a fraction α of the decomposition rate D (i.e., αD) is transferred in equal amounts to the four neighboring grid cells. Hence, in each cell microorganisms 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. Therefore, instead of modeling mobile carbon explicitly, we assumed that a fraction of the decomposition rate is transported to neighboring cells. This mass transfer mechanism can be interpreted as a consequence of various types of spatial redistribution, including diffusion or bio-turbation. The micro-scale equations at one grid cell (control volume) take the following form,

$$\frac{dC_s}{dt} = I - D_{i,j} + T_{i,j} \quad (2)$$

$$\frac{dC_b}{dt} = Y \left[(1 - \alpha) D_{i,j} + \frac{\alpha}{4} (D_{i-1,j} + D_{i+1,j} + D_{i,j-1} + D_{i,j+1}) \right] - T_{i,j} \quad (3)$$

$$\frac{dCO_{2i,j}}{dt} = (1 - Y) \left[(1 - \alpha) D_{i,j} + \frac{\alpha}{4} (D_{i-1,j} + D_{i+1,j} + D_{i,j-1} + D_{i,j+1}) \right] \quad (4)$$

where I is the rate of external input of organic C, D is the rate of decomposition, T is the microbial mortality, and Y is the microbial carbon use efficiency. The substrate C_s and microbial carbon C_b are the state variables of the micro-scale model, and their mass balances, Eq. (2) and (3), describe their temporal evolution in absence of a spatial dimension. If $\alpha = 0$, no mass transfer occurs and the model reduces to a simplified reactive system with two C pools, where grid cells are disconnected and thus independent. If $\alpha > 0$, mass transfer among the grid cells occurs. In this way, by changing the value of α , the effect of spatial redistribution on mean carbon dynamics can be assessed. With $\alpha = 0$, the general mathematical description of the simplified microscale model is given by

$$\frac{dC_s}{dt} = I - D + T, \quad (5)$$

$$\frac{dC_b}{dt} = Y D - T, \quad (6)$$

The rate of decomposition is described by three commonly used formulations: linear (Eq.7), multiplicative (Eq.8), and-MM (Eq.9) (Wutzler and Reichstein, 2008; Manzoni and Porporato, 2009), and IMM (Eq.10) (comparisons among these formulations can be found in Schimel and Weintraub (2003); Wutzler and Reichstein (2008); Manzoni and Porporato (2009)).

Table 1. Summary of the microscopic decomposition functions and steady state solutions

	Conventional (subscript L)	Multiplicative (subscript M)	Michaelis-Menten (subscript MM)	Inverse Michaelis-Menten (subscript IMM)
D	$k_L C_s$	$k_M C_s C_b$	$\frac{k_{MM} C_s C_b}{C_s + K_{MM}}$	$\frac{k_{IMM} C_s C_b}{K_{IMM} + C_b}$
T	$k_B C_b$	$k_B C_b$	$k_B C_b$	$k_B C_b$
Steady state C_s^*	$\frac{I}{(1-Y)k_L}$	$\frac{k_B}{Y k_M}$	$\frac{K_{MM} k_B}{Y k_{MM} - k_B}$	$\frac{k_B C_b}{K_{IMM} + C_b}$
Steady state C_b^*	$\frac{Y I}{(1-Y)k_B}$	$\frac{Y I}{(1-Y)k_B}$	$\frac{Y I}{(1-Y)k_B}$	$\frac{Y I}{(1-Y)k_B}$

$$D = k_{s,lin} C_s, \quad (7)$$

$$D = k_{s,mult} C_s C_b, \quad (8)$$

$$D = k_{s,mm} \frac{C_s C_b}{(K_M + C_s)} \frac{C_s C_b}{K_{MM} + C_s} \quad (9)$$

$$5 \quad D = k_{IMM} \frac{C_s C_b}{K_{IMM} + C_b}, \quad (10)$$

where $k_{s,lin}$, $k_{s,mult}$ and $k_{s,mm}$, k_L , k_M , k_{MM} , and k_{IMM} are the decomposition rate constants for linear, multiplicative, and ~~MM~~MM, and IMM kinetics respectively; and K_M is K_{MM} and K_{IMM} are the half saturation constant for the MM kinetics and IMM kinetics respectively. Table 1 summarizes the functional form-forms of D and corresponding steady state solutions for each case. Microbial mortality is typically-assumed to follow first order kinetics ($T = k_B C_b$). We assume constant temperature and soil moisture conditions so that D is only a function of C_s and C_b . This assumption facilitates assessing the role of spatial heterogeneity of C substrates and microbial biomass in our idealized system. The analytical upscaling theory developed in the following section is based on the simplified micro-scale model given by Eq. (5) and (6). For the mass-transfer model, only the numerical averaging method is used.

2.1.2 Spatial upscaling of soil carbon dynamics: scale transition theory

15 The scale transition theory links the dynamics at two different scales (Melbourne and Chesson, 2006; Morozov and Poggiale, 2012). Here we applied it is applied to study the C dynamics at micro- and macro-scale, and derive the changes in the structure of the equations describing the C pools and their fluxes at macro-scale. To upscale the micro-scale model, the spatial averaging

operator given by Eq. (1) is applied to Eq. (5) and (6), leading to the governing equations at the macro-scale,

$$\frac{d\overline{C}_s}{dt} = \overline{I} - \overline{D} + \overline{T}, \quad (11)$$

$$\frac{d\overline{C}_b}{dt} = \overline{Y}\overline{D} - \overline{T}, \quad (12)$$

where the overbars denote the spatially averaged micro-scale quantities, so that \overline{D} and \overline{T} are the macro-scale rates of decomposition and microbial mortality. Since the order of averaging and differentiation can be exchanged, the right hand side of Eq. (11) and (12) can be written as $\frac{d\overline{C}_s}{dt}$ and $\frac{d\overline{C}_b}{dt}$. Moreover, we assume that Y and I are spatially invariant, so that averaging does not alter their values. The final mass balance equations for substrate and microbial C at macro-scale are thus given by

$$\frac{d\overline{C}_s}{dt} = \overline{I} - \overline{D} + \overline{T}, \quad (13)$$

$$\frac{d\overline{C}_b}{dt} = \overline{Y}\overline{D} - \overline{T}, \quad (14)$$

$$\overline{R} = \frac{d\overline{CO_2}}{dt} = (1 - Y)\overline{D}. \quad (15)$$

where \overline{R} is the mean respiration at macro-scale. In Eq.(13)-(15), the macro-scale variables \overline{C}_s and \overline{C}_b can be obtained once the average fluxes \overline{D} and \overline{T} are known. The next step is therefore to express \overline{D} and \overline{T} as a function of ~~macro-and-micro-seale state-variables:-macro-scale~~ ($\overline{C}_s, \overline{C}_b$, and \overline{k}), and micro-scale state variables (C_s, C_b respectively).

We can generalize the problem and consider a generic microscopic C flux (i.e. D or T) as a nonlinear (and smooth) function F of state variables C_s, C_b and a parameter vector \mathbf{k} ($[k_1, k_2, \dots, k_n]$), where n is the number of parameters. The spatial averages of C_s, C_b and k are denoted as $\overline{C}_s, \overline{C}_b$ and \overline{k} ($[\overline{k}_1, \overline{k}_2, \dots, \overline{k}_n]$). Applying the averaging operator given by Eq. (1) to a multi-variate Taylor's series expansion of $F(C_s, C_b, k)$ around the spatial average value of C_s, C_b and \overline{k} and truncating the series to second order gives the macroscopic C flux (detailed derivation is provided in Appendix A1),

$$\begin{aligned} \overline{F}(C_s, C_b, \mathbf{k}) = & F(\overline{C}_s, \overline{C}_b, \overline{\mathbf{k}}) + \frac{1}{2} \frac{\partial^2 F}{\partial C_s^2} \Big|_{\overline{C}_s, \overline{C}_b, \overline{\mathbf{k}}} \sigma_{C_s}^2 + \frac{1}{2} \frac{\partial^2 F}{\partial C_b^2} \Big|_{\overline{C}_s, \overline{C}_b, \overline{\mathbf{k}}} \sigma_{C_b}^2 + \sum_{i=1}^n \sum_{j=1}^n \frac{\partial^2 F}{\partial k_i \partial k_j} \Big|_{\overline{C}_s, \overline{C}_b, \overline{\mathbf{k}}} \overline{k'_i k'_j} + \\ & \frac{\partial^2 F}{\partial C_s \partial C_b} \Big|_{\overline{C}_s, \overline{C}_b, \overline{\mathbf{k}}} \overline{C'_s C'_b} + \sum_{i=1}^n \frac{\partial^2 F}{\partial k_i \partial C_s} \Big|_{\overline{C}_s, \overline{C}_b, \overline{\mathbf{k}}} \overline{k'_i C'_s} + \sum_{i=1}^n \frac{\partial^2 F}{\partial k_i \partial C_b} \Big|_{\overline{C}_s, \overline{C}_b, \overline{\mathbf{k}}} \overline{k'_i C'_b}, \end{aligned} \quad (16)$$

where $\overline{F}(C_s, C_b, \mathbf{k})$ is the macroscopic C flux, $\sigma_{C_s}^2$ and $\sigma_{C_b}^2$ are the spatial variances of substrate and microbial C respectively; $\overline{k'_i k'_j}$ is the spatial variance (if $i = j$) or spatial covariance (if $i \neq j$) between the micro-scale parameters; $\overline{C'_s C'_b}$, $\overline{k'_i C'_s}$ and $\overline{k'_i C'_b}$

are the spatial covariances between micro-scale substrate and microbial C, substrate and parameters, and microbial biomass and parameters, respectively.

In Eq. (16), the first term on the right hand side, $F(\overline{C}_s, \overline{C}_b, \overline{\mathbf{k}})$, represents the first order approximation of $\overline{F}(C_s, C_b, \mathbf{k})$, also known as ‘mean-field’ approximation (MFA). ~~For multiplicative kinetics, MFA is given by $k_{s,mult}\overline{C}_s\overline{C}_b$ and for MM kinetics it is $k_{s,mm}\overline{C}_s\overline{C}_b/(K_M + \overline{C}_s)$~~ The MFAs for the chosen models are: $k_M\overline{C}_s\overline{C}_b$ for multiplicative kinetics; $k_{MM}\overline{C}_s\overline{C}_b/(K_{MM} + \overline{C}_s)$ for Michaelis-Menten kinetics; $k_{IMM}\overline{C}_s\overline{C}_b/(K_{IMM} + \overline{C}_b)$ for inverse Michaelis-Menten kinetics. Most C cycling models neglect all the other terms in Eq. (16). The remaining six spatial variance and covariance terms in Eq. (16) are collectively referred to as ‘second order terms’ (SOT). When the system is well-mixed, all variances and covariance terms vanish, leaving only the MFA. Therefore, only considering the MFA is equivalent to assuming well-mixed conditions at the macro-scale (i.e., Eq. (5) and (6) are mathematically equivalent to Eq. (13) and (14)).

Equation (16) provides a proof that the ‘mean-field’ approximation is a specific case of the more general expression for a macroscopic C flux that also depends on spatial heterogeneity through the SOT. The MFA is valid only when either of the following two conditions are met. First, the micro-scale decomposition rate is assumed to follow first order kinetics, because when F is a linear function of substrate and microbial C, the second order partial derivatives in Eq. (16) are zero. Second, C_s, C_b and kinetic parameters are spatially homogeneous, because in this case all the second order moments (spatial variances and covariances) are zero. However, if F is nonlinear, the second order partial derivatives are non-zero; similarly, if any type of micro-scale biophysical or biochemical heterogeneity is present, the SOT in Eq. (16) play a role in determining the macroscopic C dynamics.

Equation (16) illustrates the advantage of using scale transition theory as it provides an approximate analytical relation between the micro- and macro-scale quantities, which allows an immediate assessment of the role of both nonlinearities in the C flux formulations and spatial heterogeneities. Importantly, in some cases, Eq. (16) yields an exact (rather than approximated) equation for macro-scale quantities, as shown in the following section.

2.2 Effect of micro-scale heterogeneities on macro-scale dynamics

Depending upon the kinetics of the micro-scale decomposition model (Table 1), the macro-scale \overline{D} is expected to take different forms. Using different kinetic models, we now discuss some specific cases of micro-scale heterogeneities based on their biophysical or biochemical nature. Biophysical heterogeneity is characterized by the spatially heterogeneous distribution of substrate and microbial C, whereas biochemical heterogeneity is characterized by the spatially heterogeneous distribution of substrate quality and microbial properties, captured by the kinetic parameters. The inaccessibility of SOM can result in C

persistence. Therefore, inaccessibility can be modelled (at least at a conceptual level) through kinetic rate constants, similar to biochemical properties. In the simple model used here, accessibility to substrates or chemical recalcitrance are not mechanistically distinguished, so variations in substrate ‘quality’ in the broadest sense can be interpreted as spatial heterogeneity in either chemical characteristics or accessibility at the microscale.

- 5 First, we focus on systems with only biophysical heterogeneity of substrate and microbial C. For the first order kinetics model, the rate of decomposition is given by $\overline{D} = k_{s,lin} \overline{C_s}$ and using Eq. (16) and substituting $\overline{F} = \overline{D} = k_{s,lin} \overline{C_s}$ we obtain

$$\overline{D} = k_{s,lin} \overline{C_s}. \quad (17)$$

- In Eq. (17), \overline{D} has the same form as D , indicating that microbial-implicit first order kinetic models do not show any sensitivity to spatial heterogeneities because of the linearity of the decomposition function. For the multiplicative model, the rate of decomposition at the micro-scale is given by

$$D = k_{s,mult} C_s C_b,$$

where $k_{s,mult}$ is spatially constant and the initial values of the state variables C_s and C_b are spatially variable. Inserting Eq. (8) into Eq. (16) gives

$$15 \quad \overline{D} = k_{s,mult} \overline{C_s C_b} + k_{s,mult} \overline{C'_s C'_b}. \quad (18)$$

- In Eq. (18), the biophysical heterogeneities play a role through the covariance term $\overline{C'_s C'_b}$. Note that Eq. (18) is an exact equation because all the spatial moments of order higher than two are zero. Thus, only the mean state variables and the spatial covariance are needed to fully characterize the macro-scale dynamics for this case. Furthermore, a positive spatial covariance (i.e. co-location of substrates and microorganisms) would increase the mean decomposition rate (\overline{D}), whereas a negative spatial covariance (i.e. spatial separation between substrates and microorganisms) would decrease it.

Similar to the multiplicative decomposition model, also in models based on MM and IMM kinetics the rate of decomposition at the macro-scale depends on the covariance term $\overline{C'_s C'_b}$ and an additional term representing the spatial variance of the substrate and microbial C (Table 2). The spatial variance of the substrate term is always negative because the variance is a positive quantity and the partial derivative multiplying the variance is

derivatives multiplying the variances are negative in all decomposition functions that saturate at high substrate concentration.

In contrast, the spatial covariance term is positive or negative based on the sign of $\overline{C'_s C'_b}$. Therefore, when using the MM kinetics, \overline{D} can be approximated by the MFA only if variance and covariance balance each other or are both negligible.

Second, we consider only biochemical heterogeneity. In this case, model parameters \overline{C}_b and \overline{k} ($[\overline{k}_1, \overline{k}_2, \dots, \overline{k}_n]$) vary spatially

5 but the initial value of state variables C_s and C_b are constant everywhere in the domain. With linear decomposition, substituting

$\overline{D} = \overline{k}_{s,lin} \overline{C}_s \overline{C}_b = \overline{k}_L \overline{C}_s$ into Eq. (16) yields

$$\overline{D} = \overline{k}_{s,lin} \overline{C}_s + \overline{k}'_L \overline{C}'_s. \quad (19)$$

Equation (19) shows that for a biochemical heterogeneous system, even the simplest linear model requires an additional covariance term to describe the governing equations at the macro-scale. This covariance term might change the linear microscopic

10 model into a nonlinear macroscopic model. For the multiplicative model (Eq. (??8)), Eq. (16) yields

$$\overline{D} = \overline{k}_{s,mult} \overline{C}_s \overline{C}_b + \overline{C}_b \overline{k}'_M \overline{C}'_s + \overline{C}_s \overline{k}'_M \overline{C}'_b, \quad (20)$$

where $\overline{k}'_{s,mult} \overline{C}'_s$ and $\overline{k}'_{s,mult} \overline{C}'_b$ $\overline{k}'_M \overline{C}'_s$ and $\overline{k}'_M \overline{C}'_b$ are respectively the spatial covariances between the state variables C_s , C_b and the rate constant parameter $\overline{k}_{s,mult} \overline{k}_M$. These two additional spatial covariance terms capture the effects of biochemical heterogeneity caused by the spatial variation in the rate constants of decomposition.

15 Lastly, we consider a heterogeneous system with combined biophysical and biochemical heterogeneities, denoted as ‘fully heterogeneous’. Again, we use the multiplicative model to illustrate the relation between the dynamics at the micro- and macro-scale. Now, all the state variables and parameters in D at the micro-scale are spatially variable. For the multiplicative kinetics, $\overline{k}_{s,mult} \overline{k}_M$, C_s and C_b are spatially variable, so that inserting Eq. (??8) into Eq. (16) gives

$$\overline{D} = \overline{k}_{s,mult} \overline{C}_s \overline{C}_b + \overline{C}_b \overline{k}'_M \overline{C}'_s + \overline{C}_s \overline{k}'_M \overline{C}'_b + \overline{k}_{s,mult} \overline{C}'_s \overline{C}'_b. \quad (21)$$

20 This generalized case includes all the spatial covariances between parameters and the state variables, thereby capturing biophysical and biochemical heterogeneities simultaneously. Moreover, Eq. (20) and (21) are second order approximations, but an exact equation can be obtained by including a third order term $\overline{k}'_{s,mult} \overline{C}'_s \overline{C}'_b \overline{k}'_M \overline{C}'_s \overline{C}'_b$. A similar derivation is described for MM and IMM kinetics in the Appendix; however, an exact expression for the macro-scale MM decomposition rate for these

two kinetics cannot be found and we only use the second order approximation. Table 2 provides a summary of the theoretical results for the discussed heterogeneous cases and for all ~~three~~ four types of decomposition kinetics.

Similar derivations can be done for the microbial mortality rate (~~$F = TF = T$~~). The Taylor expansion of microbial mortality is simpler because we assume T to follow first order kinetics ~~. This implies~~ implying that all the second order terms are equal

5 to zero, ~~and \overline{T} is obtained as~~. Therefore, the mean field approximation is exact and the spatial variance of microbial C has no effect on the macro-scale dynamics.

$$\overline{T} = k_B \overline{C}_b. \quad (22)$$

~~The rate of decomposition at~~ To illustrate how macro-scale decomposition kinetics are affected by spatial heterogeneity, we define a macro-scale ~~can be used to calculate the~~ specific growth rate (SGR) ~~of the microorganisms—i.e., the respiration rate~~ divided by the ~~which is calculated by dividing the mean respiration rate by~~ mean microbial C ~~, in the system.~~

$$SGR = \frac{\overline{R}}{\overline{C}_b} = (1 - Y) \frac{\overline{D}}{\overline{C}_b}. \quad (23)$$

To summarize ~~the proposed upscaling approach~~, we started with the spatial averaging of the SOM dynamics equations at micro-scale and applied scale transition theory to derive relations between the micro- and macro-scale C fluxes, which depend on both mean state variables and their spatial statistics (Table 2). Thus, to solve the macro-scale Eq. (13) and (14), we still

15 need information regarding the second order moments i.e., $\sigma_{C_s}^2$ and $\overline{C'_s C'_b}$. To close the problem mathematically, ~~$\sigma_{C_s}^2$ and $\overline{C'_s C'_b}$~~ these moments can be regarded as extra state variables requiring additional differential equations describing their dynamics (Keeling et al., 2002; Murrell et al., 2004; Barraquand and Murrell, 2013). Alternatively, the second order ~~terms~~ moments can be parameterized as empirical functions of first order terms \overline{C}_s , \overline{C}_b and \overline{k} (Bergström et al., 2006). Here, our goal is to quantify how heterogeneities alter C fluxes in idealized systems, so we leave the closure problem for a future contribution and

20 use instead the numerically simulated dynamics at the micro-scale to calculate the spatial moments ~~$\sigma_{C_s}^2$ and $\overline{C'_s C'_b}$~~ and SOT.

Table 2. Summary of macro-scale equations for the decomposition rate (\bar{D})

	Biophysical heterogeneity	Biochemical heterogeneity	Full-heterogeneity
Linear	$\cancel{k_{s,lin} \bar{C}_s} \cancel{k_L \bar{C}_s}$	$\bar{k}_{s,lin} \bar{C}_s + \bar{k}'_{s,lin} \bar{C}'_s \bar{k}_L \bar{C}_s + \bar{k}'_L \bar{C}'_s$	$\bar{k}_{s,lin} \bar{C}_s + \bar{k}'_{s,lin} \bar{C}'_s \bar{k}_L \bar{C}_s + \bar{k}'_L \bar{C}'_s$
Multiplicative*	$\frac{k_{s,mult} \bar{C}_s \bar{C}_b}{+ k_{s,mult} \bar{C}'_s \bar{C}'_b} \frac{k_M \bar{C}_s \bar{C}_b}{+ k_M \bar{C}'_s \bar{C}'_b}$	$\frac{k_{s,mult} \bar{C}_s \bar{C}_b}{+ \bar{C}_s \bar{k}'_{s,mult} \bar{C}'_b + \bar{C}_s \bar{k}'_M \bar{C}'_b} \frac{k_M \bar{C}_s \bar{C}_b}{+ \bar{C}_b \bar{k}'_{s,mult} \bar{C}'_s + \bar{C}_b \bar{k}'_M \bar{C}'_s}$	$\frac{k_{s,mult} \bar{C}_s \bar{C}_b}{+ \bar{C}_s \bar{k}'_{s,mult} \bar{C}'_b + \bar{C}_s \bar{k}'_M \bar{C}'_b} \frac{k_M \bar{C}_s \bar{C}_b}{+ \bar{C}_b \bar{k}'_{s,mult} \bar{C}'_s + \bar{C}_b \bar{k}'_M \bar{C}'_s}$
Michaelis-Menten	$\frac{k_{s,mm} \bar{C}_s \bar{C}_b}{K_M + \bar{C}_s} + \frac{1}{2} \left[\frac{-2k_{s,mm} K_M \bar{C}_b}{K_M + \bar{C}_s^2} \right] \sigma_{\bar{C}_s}^2 + \frac{k_{MM} \bar{C}_s \bar{C}_b}{K_{MM} + \bar{C}_s} + \frac{1}{2} \left[\frac{-2k_{MM} K_{MM} \bar{C}_b}{(K_{MM} + \bar{C}_s)^3} \right] \sigma_{\bar{C}_s}^2 +$ $\left[\frac{k_{s,mm} K_M}{(\bar{C}_s + K_M)^2} \right] \bar{C}'_s \bar{C}'_b \quad \left[\frac{k_{MM} K_{MM}}{(\bar{C}_s + K_{MM})^2} \right] \bar{C}'_s \bar{C}'_b$	Eq.(A8)	Eq.(A9)
Inverse Michaelis-Menten	$\frac{k_{IMM} \bar{C}_s \bar{C}_b}{K_{IMM} + \bar{C}_b} + \frac{1}{2} \left[\frac{-2k_{IMM} K_{IMM} \bar{C}_b}{(K_{IMM} + \bar{C}_b)^3} \right] \sigma_{\bar{C}_b}^2 +$ $\left[\frac{k_{IMM} K_{IMM}}{(\bar{C}_b + K_{IMM})^2} \right] \bar{C}'_s \bar{C}'_b$		

* The expression of \bar{D} for multiplicative kinetics in each heterogeneity case is exact.

~~The expression of \bar{D} for multiplicative kinetics in each heterogeneity case is exact.~~

2.3 Model setup

To investigate the effect of micro-scale spatial heterogeneity of SOM on the decomposition dynamics, we use a synthetic approach, which allows considering a wide range of heterogeneous SOM fields. As in other [As in previous](#) spatially explicit models (Ginovart and Valls, 1996; Allison, 2005; Kaiser et al., 2014), we start with a 2D domain characterized by an initial heterogeneous field of the substrate and numerically simulate the dynamics of SOM with the micro-scale two pool model in Eq. (5) and (6) at each cell in the domain. The 2D domain has 100×100 square grid cells with an edge length of $50 \mu\text{m}$, and we populate it with randomly generated initial substrate fields. This numerical model is referred to as ‘distributed model’ (see, Fig. 1). From the solution of the distributed model, the mean behavior of the system ($\overline{C_s}, \overline{C_b}, \overline{D}, \sigma_{C_s}^2, \overline{C_s}, \overline{C_b}, \overline{D}, \sigma_{C_s}^2, \sigma_{C_b}^2$, and $\overline{C'_s C'_b}$) can be calculated at each time step by using sample statistics of C_s and C_b

$$\overline{C_s}(t) \approx \frac{1}{N_x N_y} \sum_{i=1}^{N_x} \sum_{j=1}^{N_y} C_{s,i,j}(t), \quad (24)$$

$$\overline{C_b}(t) \approx \frac{1}{N_x N_y} \sum_{i=1}^{N_x} \sum_{j=1}^{N_y} C_{b,i,j}(t), \quad (25)$$

$$\overline{D}(t) \approx \frac{1}{N_x N_y} \sum_{i=1}^{N_x} \sum_{j=1}^{N_y} k_{s,mult,i,j} M_{i,j} C_{s,i,j}(t) C_{b,i,j}(t), \quad (26)$$

$$\sigma_{C_s}^2(t) \approx \frac{1}{N_x N_y} \sum_{i=1}^{N_x} \sum_{j=1}^{N_y} [C_{s,i,j}(t) - \overline{C_s}(t)]^2, \quad (27)$$

$$\sigma_{C_b}^2(t) \approx \frac{1}{N_x N_y} \sum_{i=1}^{N_x} \sum_{j=1}^{N_y} [C_{b,i,j}(t) - \overline{C_b}(t)]^2, \quad (28)$$

$$\overline{C'_s C'_b}(t) \approx \frac{1}{N_x N_y} \sum_{i=1}^{N_x} \sum_{j=1}^{N_y} [C_{s,i,j}(t) - \overline{C_s}(t)] [C_{b,i,j}(t) - \overline{C_b}(t)], \quad (29)$$

where \overline{D} is specified for multiplicative kinetics; a similar approach was applied for MM kinetics. Table B1 in the appendix lists all the parameters related to different kinetic models used in simulations. We performed the simulation in mass units fg (fg = 10^{-15} g) and later converted the state variables to concentration units i.e. mgC/g of soil.

2.4 Initial 2D random fields of SOM and kinetic parameters

Two-dimensional spatially ~~correlated random fields~~ [heterogeneous distributions](#) of substrates and microbial C were generated to run the distributed model. ~~For the heterogeneous field of microbial C, we used a random field generator that provides~~

100 × 100 spatially correlated random numbers between -1 and 1 (Lennon, 2000). These values were then re-scaled by an appropriate mean and standard deviation of microbial C. To simulate the dead-zones in the heterogeneous system some grid cells were forced to have no microbial C (the obtained field is denoted $y_{i,j}$). Moreover, to allow comparison among simulations, the microbial C field was re-normalized to have a specified value of total initial microbial C,

$$5 \quad C_{b,i,j} = \frac{y_{i,j}}{\sum_{i,j} y_{i,j}} C_{b,total} \text{ (fg)}$$

It is assumed that the $C_{b,total}$ is equal to 1% of the total amount of substrate in the domain (Witter, 1996), which is in turn calculated as $C_{s,total} = \bar{C}_{s,0} \times N_x \times N_y$ where $\bar{C}_{s,0}$ is the initial mean substrate C in a single grid cell (). The amount of substrate C in any grid cell is limited by the maximum amount of C that the cell can accommodate, according to the density of organic matter (ρ_{SOM}). We assume that each cell can contain between zero and 50% of this maximum amount of organic C because approximately only 50% of organic matter on a mass basis is composed of organic C. The maximum amount of substrate C that one cell can contain is thus given by

$$C_{max} = 0.5 \rho_{SOM} \text{ cell}_{volume} \text{ (fg)}$$

where cell_{volume} is the volume of a cell in grid. The value of $\bar{C}_{s,0}$ was chosen so that the maximum C amount at a micro-site does not exceed C_{max} . To summarize, the obtained spatially heterogeneous random fields of microbial C and substrate C satisfy the The obtained distributions were based on following constraints: i) the total amount of organic C is set, ii) the total amount of microbial C is 1% of total organic C, iii) the maximum amount of C in a cell is set (Eq. (A12)), and iv) some grid cells have no microbial biomass. For details regarding the procedure to generate the microbial C field, see Appendix A2.

To study the effects of degree of heterogeneity on C decomposition, ~~we generated~~ random fields of substrate C with different degrees of correlation with microbial C were generated. We created three cases where substrate and microbial C were initially positively correlated, negatively correlated or uncorrelated. The three cases were obtained by applying a linear operator on the microbial C fields with positive and negative slope to obtain positively and negatively correlated substrate C fields, respectively. The uncorrelated substrate C field was generated independently from the microbial C field and can be interpreted as the result of external disturbances disrupting preexisting spatial correlations. The case of positive initial correlation between substrate and microbial C would result in a heterogeneous system with spatial co-occurrence of substrate and microbial C, whereas initial negative correlation would result in isolated patches of substrate and microbial C. ~~The uncorrelated initial field of substrate and microbial C can be interpreted as the result of external disturbances disrupting preexisting spatial correlations.~~ In all

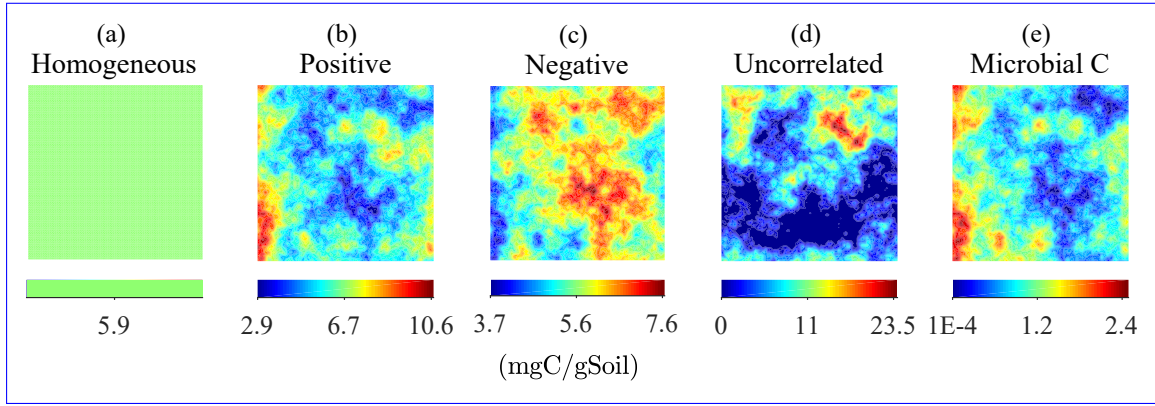


Figure 2. Steady state simulation: Examples of the homogeneous (a) and the heterogeneous distributions of substrate C constrained to have the same amount of total C-substrate (b–d). The fields in b–d were obtained by imposing different degrees of correlation of substrate C with the initial heterogeneous distributions of microbial C, as indicated above each panel shown in (e).

scenarios, the substrate distributions are normalized to have the same amount of $C_{s,total}$, thereby allowing for comparisons among different degrees of heterogeneity. Example of the heterogeneous fields of C-substrate are shown in Figure ?? (Figure 2).

To generate a heterogeneous random field for kinetic parameters, we considered a uniform distribution for K_M and a log-uniform distribution for $k_{s,mm}$ and $k_{s,mult}$ k_M , k_{MM} , and k_{IMM} (Forney and Rothman, 2012; Manzoni et al., 2012). The log-uniform distributions were defined so that the mean kinetic constants were equal to those of the homogeneous system (Table B1) and their variances were tuned to characterize different degrees of heterogeneity. To generate the random fields, $N_x \times N_y$ random numbers were extracted from the chosen distributions and placed into a the 2D matrix of size $N_x \times N_y$ domain. Figure A7 in the appendix shows the probability densities for two different standard deviations for $k_{s,mm}$ and $k_{s,mult}$ k_{MM} and k_M (the parameters of the distributions are listed in Tables B2 and B3).

2.5 Estimation of kinetic parameters

To choose parameter values for the linear and multiplicative kinetics that allow comparisons with the MM model, we first simulated the substrate C dynamics at micro-scale for a given initial condition and using MM kinetics. Second, we fit the linear and multiplicative kinetics models to the time series obtained using MM kinetics (using the optimization toolbox in MATLAB). In this procedure, we assumed that the microbial mortality constant (k_B) was the same for all choices of the decomposition model. The parameters of the inverse MM are chosen (by trial and error) so that the respiration rate in the homogeneous system is comparable to that in the heterogeneous system.

2.6 Simulation scenarios

Two scenarios (Fig. 3), based on varying initial conditions (IC) were implemented to investigate the effects of micro-scale heterogeneities on macroscopic decomposition.

Scenario 1 (steady state ~~initial conditions~~simulation - SS): In this scenario, the initial heterogeneous field of substrate and microbial C was generated as described in section 2.4. The spatial mean of the initial substrate and microbial C match their steady state values given by the micro-scale equations (Eq. (5) and (6)) forced with a constant substrate input. Additionally, ~~in the dead zones a small amount (i.e.,~~ a minimum amount of microbial C was set in each cell (values at least one order of magnitude ~~ranging from one to two less than the~~ lower than those at steady state) ~~of microbial C was imposed~~ to ensure that OM could be decomposed, ~~even if much slower~~ albeit at a slower rate than elsewhere.

Scenario 2 (~~transient initial condition~~high substrate simulation - HS): In the scenario, the initial heterogeneous field of substrate C was perturbed around a value much larger than the steady state as described in section 2.4.

In the ~~transient IC~~HS scenario, simulations were based on ~~two~~three nonlinear decomposition models (multiplicative~~and MMkinetic, MM, and IMM kinetics~~). However, in the ~~steady state IC~~SS scenario, we present results only for multiplicative kinetics because MM kinetics can be approximated by multiplicative kinetics when the substrate is much smaller than the half-saturation constant ($K_M K_{MM}$), as is the case with the chosen initial heterogeneous substrate field and the parameter $K_M K_{MM}$. Results using the linear decomposition model are not shown because with this model the spatially-averaged fluxes are equal to the macro-scale flux calculated at the mean C concentration ~~-(Eq. (17)).~~

In both scenarios, we explore the effects of biophysical and full heterogeneity on the temporal evolution of the mean state variables (substrate and microbial C) and their ~~fluxes~~associated rates. We used the distributed ~~modeling approach~~model to estimate the mean quantities and second order spatial moments (and thus SOT) for three degrees of biophysical heterogeneity. A homogeneous system in which the initial substrate and microbial C, as well as kinetic parameters, are spatially uniform was always used as a control. The combined effect of biophysical and biochemical heterogeneity was simulated by imposing the spatially heterogeneous kinetic parameters along with the heterogeneous initial substrate and microbial C.

3 Results

3.1 Scenario 1: ~~Steady steady~~ state ~~initial conditions~~simulation (SS)

Figure 4 illustrates the temporal evolution of the macroscopic decomposition dynamics for the three different heterogeneous cases with varying degrees of initial correlation between substrate and microbial C in comparison to the homogeneous system.

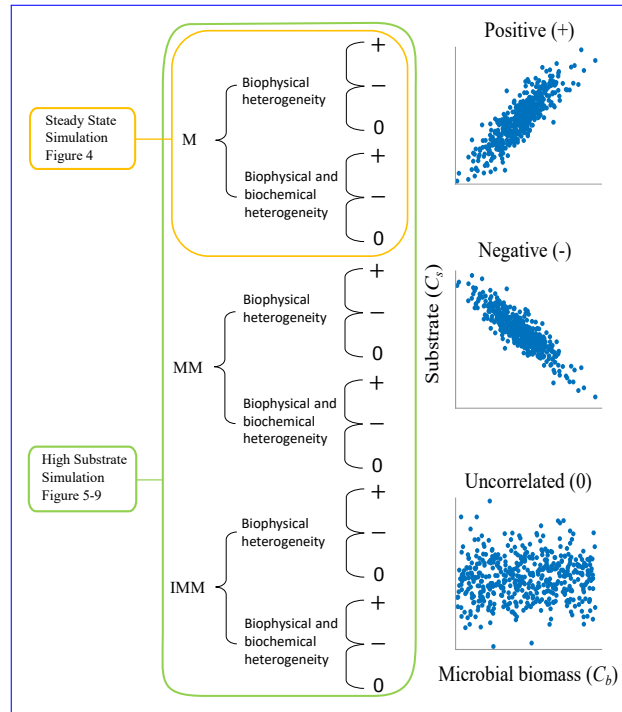


Figure 3. Two scenarios were implemented based on initial spatial distribution of substrate and microbial C. In scenario 1, substrate and microbial C are perturbed around the steady state of the micro-scale differential equations and simulations are only carried out with the multiplicative (M) kinetics. In scenario 2, substrate and microbial C are perturbed to values larger than the steady state, and simulations are conducted for both multiplicative (M), Michaelis-Menten (MM) and inverse Michaelis-Menten (IMM) kinetics. For each scenario and type of heterogeneity, three different initial distributions of substrate and microbial biomass are considered as representative of micro-scale heterogeneities (positively correlated (+), negatively correlated (-), and uncorrelated (0) fields of substrate and microbial C).

In figure 4, the left and right panels respectively show the effects of biophysical heterogeneity and full heterogeneity on the mean C pools and fluxes ($k_{s,mult} k_M$ is based on the case 'biochemical heterogeneity 1' in Table B3). For this analysis, we focus on the multiplicative decomposition model.

Since the mean initial condition corresponds to the steady state of the micro-scale system, in a homogeneous soil no changes occur in substrate C (solid line in Fig. 4a and 4eb) and microbial C (solid line in Fig. 4b and 4fd), and the mean respiration rate is equal to the constant rate of input of external C (solid line in Fig. 4e and 4ge and 4f). In contrast, for the system with biophysical heterogeneity, the mean C pools and respiration (\bar{R}) fluctuate towards the steady state of the micro-scale system as a result of the heterogeneous initial placement of C substrates. Similarly, for the fully heterogeneous system the mean microbial C pool (Fig. 4fd) and fluxes (Fig. 4gf) fluctuate near their steady state values, but the mean substrate C pool (Fig. 4eb) reaches a new steady state. The value of the new steady state for the \bar{C}_s depends upon the parameters of the log-uniform distribution of $k_{s,mult} k_M$ and is given by $[k_B(10^{-a} - 10^{-b})/Y(b-a)\ln(10)]$ (see details in the Appendix) the Appendix

A3. In all heterogeneity scenarios, \bar{R} is initially higher than in the homogeneous system when substrate and microbial C are initially correlated, whereas it is lower when substrate and microbial C are negatively correlated. When substrate and microbial C are uncorrelated, the system exhibits a behavior similar to that of the positively correlated fields (Fig. 4ee), but with higher respiration peaks. This is caused by the high initial spatial variance of substrate C that resulted in hot spots richer in substrate C than in the positively correlated case (Fig. 2). Furthermore, in the multiplicative kinetics, the respiration flux is proportional to the amount of substrate C, so that larger variations in substrate cause larger fluctuations in the mean respiration flux. In the fully heterogeneous system, fluxes show similar dynamics as those in the biophysically heterogeneous system, except for the different steady state. Varying the mean (Fig. A1) and variability (Fig. A2) of k_M alters the quantitative, but not qualitative, behavior of the macro-scale system (results shown in Appendix A4).

Figure 4d-g and 4h show the sum of all higher order terms ($\sum HOT$), including the third order term $\overline{k'_{s,mult} C'_s C'_b k'_M C'_s C'_b}$ in addition to the SOT. For a biophysically heterogeneous system, the $\sum HOT$ only includes the spatial covariance term, but for a fully heterogeneous system it includes the last three terms of the Eq. (21). The $\sum HOT$ is initially positive, zero and negative, respectively for positively correlated, uncorrelated, and negatively correlated substrate and microbial C. ~~All components of HOT show a dynamic behavior, and exhibits strong temporal variations~~ (Appendix, Fig. A3). A positive $\sum HOT$ value enhances \bar{R} , whereas a negative value decreases it in all three heterogeneous cases compared to homogeneous \bar{R} . This result is aligned with our expectation from the analytical expression of the macro-scale multiplicative model (Eq.18).

~~Figure ?? compares the specific growth rate (SGR) of heterogeneous and homogeneous systems as a function the mean substrate C. In the case of a homogeneous system, SGR is equal to the time-invariant ratio $\frac{R}{C_b}$ (shown as a black dot), whereas in the heterogeneous system SGR is given by~~ In systems including both biophysical and full heterogeneity, the sums of the time-dependent ratio $\frac{\bar{R}}{C_b}$. As a result, the oscillations in (\bar{R}) caused by the HOT give rise to a non-unique relation between SGR and mean substrate C. HOT are stable in the long term, once the steady state has been reached. This was confirmed by running the model for 100 years. Furthermore, any additional perturbation of the new steady state caused by an external factor will re-introduce the fluctuations.

~~Scenario 1 (initial condition at steady state): effects of biophysical and full heterogeneity on the mean specific growth rate (SGR) as a function of mean substrate C for the three heterogeneous cases and the black dot showing the value of $\frac{R}{C_b}$. The direction of the arrows represent the increasing time.~~

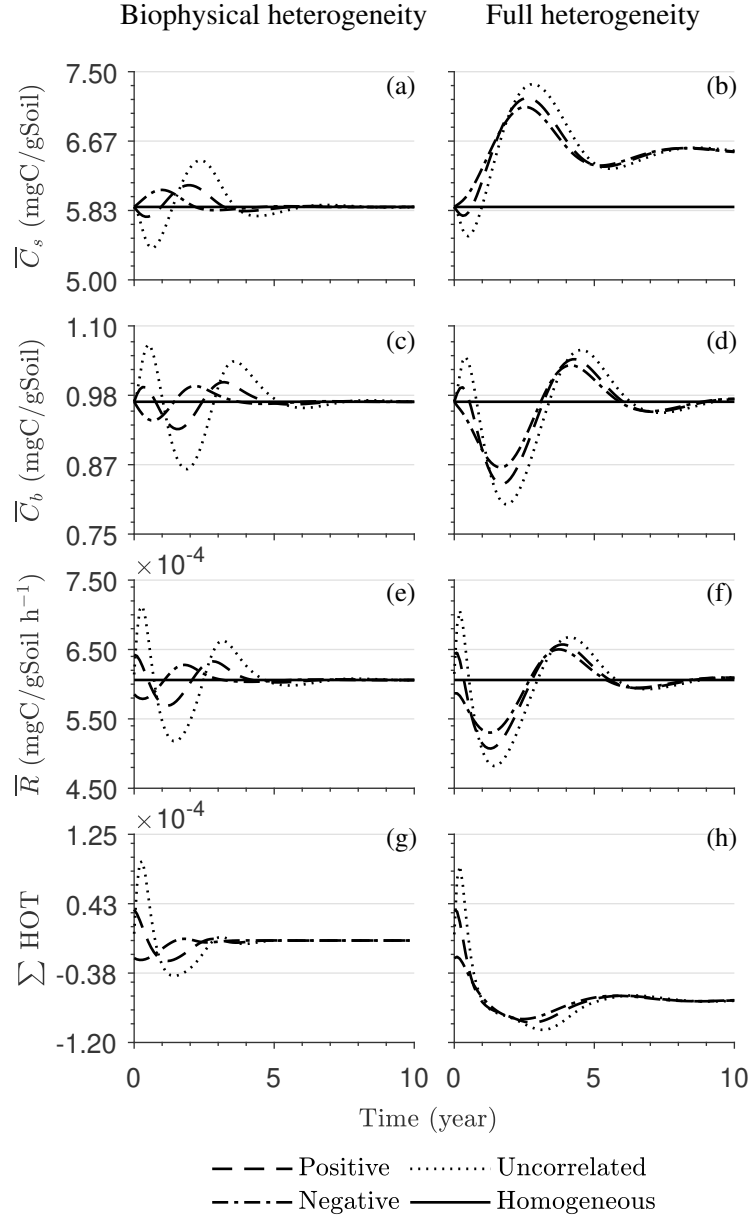


Figure 4. Scenario 1 (~~initial condition at steady state~~ [simulation](#)): effect of biophysical (left panel) and full heterogeneity (right panel) on the macroscopic decomposition dynamics when the substrate is distributed randomly around the steady state, [and only considering multiplicative kinetics at the micro-scale](#): (a,e) mean substrate C (\bar{C}_s), (b,c,f,d) mean microbial C (\bar{C}_b), (e,e,g,f) mean respiration rate (\bar{R}), and (d,g,h) sum of second and third order terms ($\sum HOT$).

3.2 Scenario 2: ~~Transient initial condition~~high substrate simulation (HS)

3.2.1 Dynamics of substrate and microbial C at the macro-scale

Figure 5 illustrates the temporal evolution of the mean properties of the macroscopic decomposition dynamics for multiplicative kinetics, for systems with either biophysical (left panel) or full (right panel) heterogeneity, when the initial condition is perturbed from the steady state by adding C substrates. In this scenario, both homogeneous and heterogeneous systems exhibit transient dynamics because the initial conditions are set far from the steady state. ~~Figure 5e and 5f show the mean respiration rate \bar{R} for the biophysically and the fully heterogeneous system, respectively.~~ The results in Fig. 5b-c indicate that, during the microbial growth phase, the production of microbial C is faster when substrate and microbial C are positively correlated or uncorrelated, compared to the case of negative correlation. Consequently, at the beginning of the simulation, the mean substrate \bar{C}_s (Fig. 5a) is decomposed faster due to the higher respiration (Fig. 5ee) for the ~~uncorrelated and the positively correlated~~ positively correlated and the uncorrelated substrate and microbial C, and slower for the negatively correlated substrate and microbial C, when compared to the homogeneous system. By the end of the simulation period, in all heterogeneous scenarios, biomass production and substrate consumption are lower than in the homogeneous system. As in scenario 1, the initial mean respiration for the uncorrelated case is higher than that in the positively correlated case.

~~The~~ Moreover, the fully heterogeneous system (Fig. 5 right panel) shows a similar behavior ~~compared to as~~ the biophysically heterogeneous system, but the peaks of \bar{R} appear earlier for all degrees of correlation between substrate and microbial C.

~~Figure 6 shows~~ Similar to Fig. 4, Fig. 5g and 5h show the sum of all higher order terms. For both heterogeneous systems, \bar{R} is higher than the MFA when the $\sum HOT$ is positive, whereas \bar{R} is lower than the MFA when the contribution of these spatial moments is negative. This result agrees with the analytical expression and holds for all types of biophysical heterogeneities.

~~The $\sum HOT$ for biophysical heterogeneity is initially positive when substrate and microbial C are positively correlated or uncorrelated, but later becoming negative, whereas it is always negative for the negatively correlated substrate and microbial C. Spatial covariances among kinetic parameter and state variables (i.e., $\overline{k_M' C_s'}$, $\overline{k_M' C_b'}$, and $\overline{k_M' C_s' C_b'}$) also contribute to the $\sum HOT$ in the fully heterogeneous system in addition to $\overline{C_s' C_b'}$. Specifically, the spatial covariance between k_M and C_b gives rise to early peaks of \bar{R} (see all HOT in Fig. A4).~~

~~Figures 6 and 7 show~~ similar results as Fig. 5, but for Michaelis-Menten kinetics and inverse Michaelis-Menten kinetics, respectively. The transient dynamics of the mean C pools and fluxes differ from those ~~for obtained using~~ multiplicative kinetics. ~~During For both MM and IMM kinetics, during~~ the initial growth period, the ~~decomposition of mean substrate C in heterogeneous systems (Fig. 6a and 6d) occurs at a rate comparable to that in the~~ mean respiration rate in the biophysically

heterogeneous systems is similar to that occurring in a homogeneous system, but afterward \bar{R} becomes higher for positive, similar for uncorrelated, and lower for negatively correlated substrate and microbial C (Fig. 6e and 7e). As a result, substrate loss (Figs. 6a and 7a) and microbial growth (Figs. 6c and 7c) slow down compared to homogeneous conditions. Interestingly, for MM kinetics with MM kinetics, in the uncorrelated case \bar{R} compared to the positively correlated case (Fig. 6e), because now is not higher than in the other heterogeneity cases as occurred with multiplicative kinetics (compare Figs. 6e and 5e). This is because with MM kinetics the respiration flux is limited by the maximum rate of decomposition and not only by substrate availability. In contrast, with IMM kinetics \bar{R} in the uncorrelated case is higher than in the other heterogeneity cases, as it was with multiplicative kinetics. This is because the initial microbial C is often much lower than the half saturation constant for IMM kinetics, making the IMM decomposition rate equation numerically similar to the multiplicative decomposition model.

The fully heterogeneous system (right panels in Fig. 5 and 6 right panel, 6, and 7) shows different behavior compared to the biophysically heterogeneous system. The peaks of \bar{R} appear much earlier than in the biophysically heterogeneous system. Additionally, the values of mean fluxes and C pools after the peak are smaller than in the homogeneous system as well as in the system with only biophysical heterogeneity. The inverse MM kinetics (Fig. 7 right panel) show similar dynamics as in the case of biophysical heterogeneity, but with reduced peak magnitude. The smaller mean fluxes are due to the left skewed probability distribution of the kinetic parameters ($k_{s,mult}$ and $k_{s,mm}$ k_M and k_{MM}), which causes slower decay despite the mean values of the kinetic parameters being the same. Mathematically, this behavior is caused by the additional covariances in the fully heterogeneous system as explained in the following paragraph.

Figure A4 presents all the higher-order spatial moments in the analytical expression of the macroscopic mean respiration rate, for the multiplicative decomposition model. The left (respectively right) vertical column shows the results for biophysically (fully) heterogeneous system with horizontal rows corresponding to the cases of positive (i.e. Fig. A4a and d), negative (i.e. Fig. A4b and e) and uncorrelated (i.e. Fig. A4c and f) substrate and biomass C. For both heterogeneous systems, \bar{R} is higher than the MFA when the $\sum HOT$ is positive, whereas \bar{R} is lower than the MFA when the contribution of these moments is negative. This result agrees with the analytical expression and are valid for all types of biophysical heterogeneities. All the higher-order spatial moments show a dynamic behavior. The $\sum HOT$ for biophysical heterogeneity is initially positive for the positively and the uncorrelated substrate and microbial C, but later becoming negative while it is always negative for the negatively correlated substrate and microbial C. Spatial covariances among kinetic parameter and state variables (i.e., $\overline{k'_{s,mult}C'_s}, \overline{k'_{s,mult}C'_b}, \overline{k'_{s,mult}C'_sC'_b}$) also contribute to the $\sum HOT$ in the fully heterogeneous system in addition to $\overline{C'_sC'_b}$. Specifically, it is the spatial covariance between $k_{s,mult}$ and C_b that gives rise to early peaks of \bar{R} .

3.2.2 Dynamics of the second order terms

The $\sum SOT$ (same as $\sum HOT$ but now limiting the HOTs to second order) for MM and IMM kinetics for the biophysical heterogeneity-biophysically heterogeneous system is given by the sum of the last two terms of Eq. \bar{D} in Table 2 and for the fully heterogeneous system it is given by the last eight terms of Eq. (A9) and Eq. (A10), respectively. For the biophysically heterogeneous system, the values of $\sum SOT$ (Appendix, Fig. A5 Fig. 6g and 7g) are initially positive (very small in magnitude) for the positively correlated substrate and microbial C and later become negative, while for other heterogeneous cases the negatively correlated heterogeneous case $\sum SOT$ is always negative. For uncorrelated substrate and microbial C, $\sum SOT$ is initially negative in MM kinetics but positive in IMM kinetics and later becomes negative. Furthermore, the balance between variance and covariance terms makes the MFA a good approximation of \bar{R} only when the combined second order terms are negligible, which is not the case in this example (see, Fig. A5 and A6). The $\sum SOT$ of the fully heterogeneous system for MM kinetics is shown in appendix Fig. ??, and values were positive for the first 100 days of simulation and then negative onward, even though the heterogeneous \bar{R} is smaller than the homogeneous \bar{R} (Fig. 6). This result suggests that for a fully heterogeneous systems the SOT approximation of the macro-scale differential equation is not sufficient and additional higher order term can not be ignored. Individual components of $\sum SOT$ are not presented in Fig. ?? because of the large number of SOTs involved. h and 7h).

3.2.3 Emerging macroscopic kinetics

Figure 8 highlights the effect of these additional SOTs (HOTs for multiplicative) effects of spatial heterogeneities on the mean specific growth rate (SGR) of the heterogeneous system for multiplicative (top) and MM (bottom), MM (middle), and IMM kinetics (bottom panels). The depicted SGR curves can be interpreted as the macroscopic kinetic laws emerging from the spatial averaging. For all three kinetics, For both kinetics, the functional relation between the mean SGR and \bar{C}_s for the heterogeneous system depends upon the initial degree of heterogeneity. In contrast, in the homogeneous system the mean SGR is a linear and saturating function of \bar{C}_s , saturating, and exponentially increasing function of C_s for the multiplicative and MM, MM, and IMM kinetics, respectively. The effect of biophysical heterogeneity in both kinetic models is all kinetic models are shown in Fig. 8a and 8e, 8c and 8e. The negative correlation between substrate and microbial C leads to lower SGR than in the homogeneous system, even if both heterogeneous and homogeneous systems have exactly the same amount of total initial substrate and microbial C. In the case of positive correlation, the initial mean SGR is higher than in the homogeneous system, but in the later phase of decomposition, mean SGR becomes lower. Thus, when the substrate is physically accessible to co-located with the microorganisms, the mean SGR is initially higher but it decreases at a faster rate as the substrate is

decomposed when compared to the homogeneous system. If substrate and microbial C are uncorrelated, the SGR functional response remains between the negative and the positive correlation cases.

In the fully heterogeneous system (Fig. 8b and 8d, 8d, and 8f), the nonlinear character of the relation between mean SGR and \overline{C}_s increases compared to the biophysically heterogeneous system. Interestingly, the mean SGR in the case of negative correlation for ~~both~~ both-multiplicative and MM kinetic models is higher (for high \overline{C}_s) than for the homogeneous system, despite being the co-location of substrates and microorganisms ~~is~~ less likely. This behavior is caused by the occurrence of patches with high turnover rate that control the mean SGR (Fig. 8a and 8c). In contrast, for IMM kinetics, the mean SGR in the case of negative and positive correlation is lower than the homogeneous system. This behavior might be a consequence of the chosen value of K_{IMM} ; i.e., in our parameterization of the IMM kinetics, initially the system is limited by microbial C, resulting in relatively low decomposition rate and dynamics comparable to those obtained with a multiplicative model (see, Fig. 7e-f).

In Fig. 9 the role of C transfer among cell is investigated. Similar to Fig. 8, we show the specific growth rate as a function of substrate for an uncorrelated initial distribution of substrate and microbial C, and for all three kinetics– multiplicative, Michaelis-Menten and inverse Michaelis-Menten. When $\alpha = 0$, result in Fig. 9 are same as in Fig. 8 for the uncorrelated case. When $\alpha > 0$, microorganism that were initially deprived of substrate now receive additional substrate from neighboring grid cells. As a consequence of this improved substrate availability, in the long-term microorganisms can consume all the substrate, whereas without mass transfer some C remains undecomposed (Figs. 5b, 6b, and 7b).

4 Discussion

4.1 Predicted effects of spatial heterogeneity on decomposition

The heterogeneous spatial distribution of organic matter in soils is a result of complex physical, chemical, and biological processes. Both the experimental quantification of the effects of heterogeneity on SOM dynamics (Kravchenko and Guber, 2017), and capturing such effects in mathematical models (Wieder et al., 2015) ~~are~~ is challenging. Here, we used scale transition theory, applied to a two pool model, as a simple approach to analytically account for spatial heterogeneities. ~~This approach was then used to~~ and upscale SOM dynamics in ~~a range of~~ idealized scenarios that cover different types of spatial heterogeneity. Even with the simplest scenarios, the macroscopic decomposition dynamics of a heterogeneous system differ from those predicted from the mean-field approximation (equivalent to assuming well-mixed conditions). This difference in the dynamics at two spatial scales arises because spatial averaging of the nonlinear kinetics at the micro-scale create additional terms in

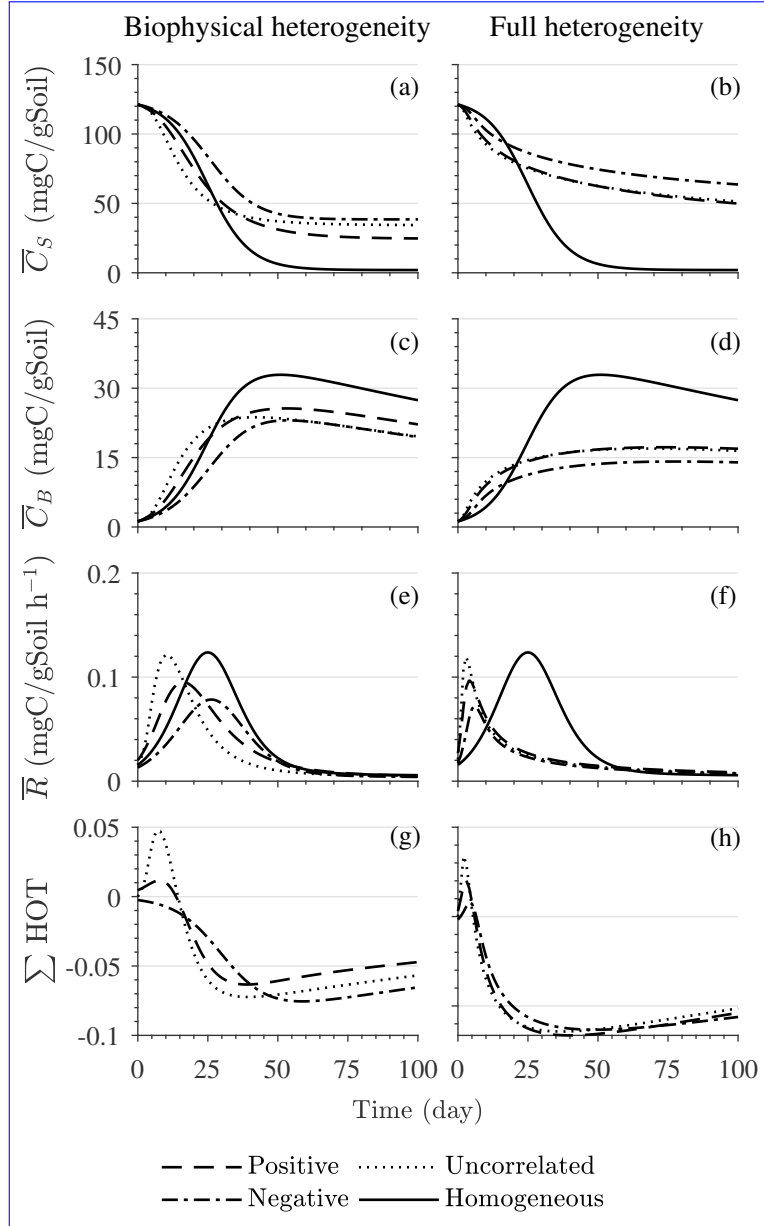


Figure 5. Scenario 2 (transient dynamics HS with multiplicative kinetics): effect of biophysical heterogeneity (left panel) and full heterogeneity (right panel) on the macroscopic decomposition dynamics when the substrate is distributed around a value higher than the steady state of the homogeneous system: (a,e) mean substrate C (\bar{C}_s), (b,c,f,d) mean microbial C (\bar{C}_b), and (e,e,g,f) mean respiration rate (\bar{R}), and (g,h) sum of second and third order terms ($\sum HOT$).

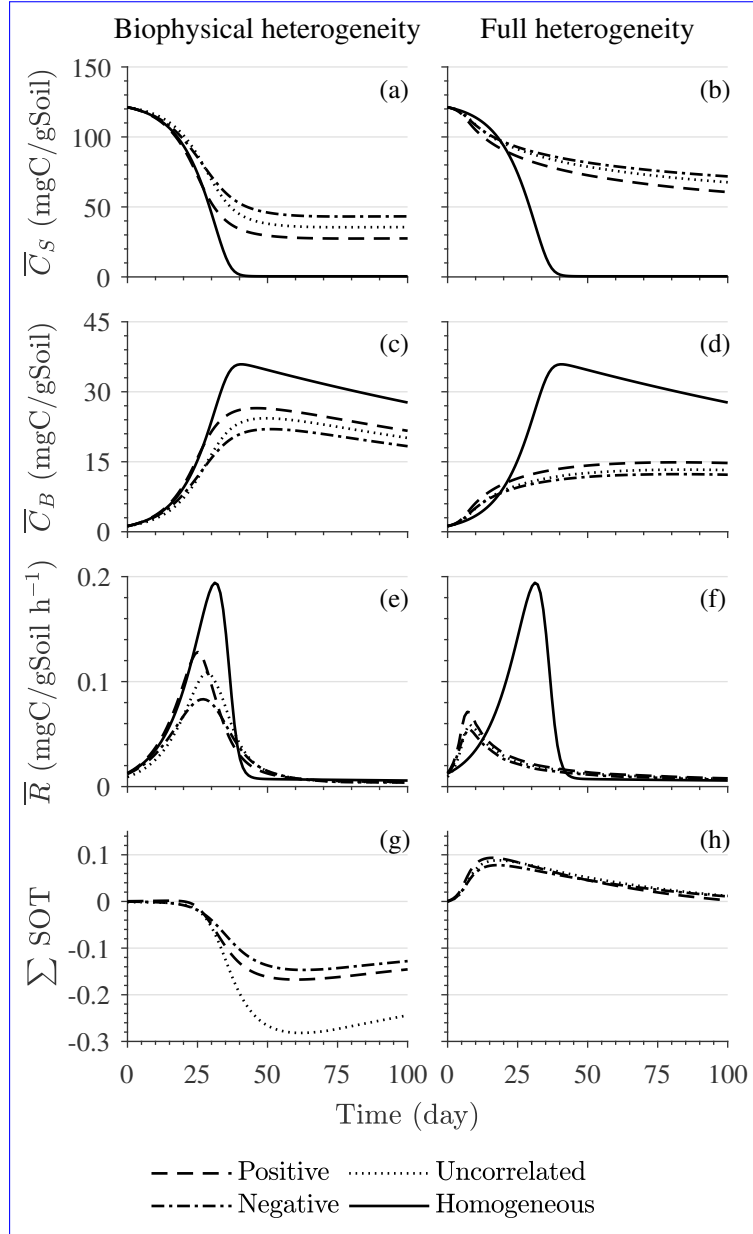


Figure 6. Scenario 2 (transient-dynamics HS with Michaelis-Menten kinetics): effect of biophysical heterogeneity (left panel) and full heterogeneity (right panel) on the macroscopic decomposition dynamics when the substrate is distributed around a value higher than the steady state of the homogeneous system: (a,e) mean substrate C (\bar{C}_s), (b,f) mean microbial C (\bar{C}_b), and (c,g) mean respiration rate (\bar{R}), and (d,h) sum of second order terms ($\sum HOT$).

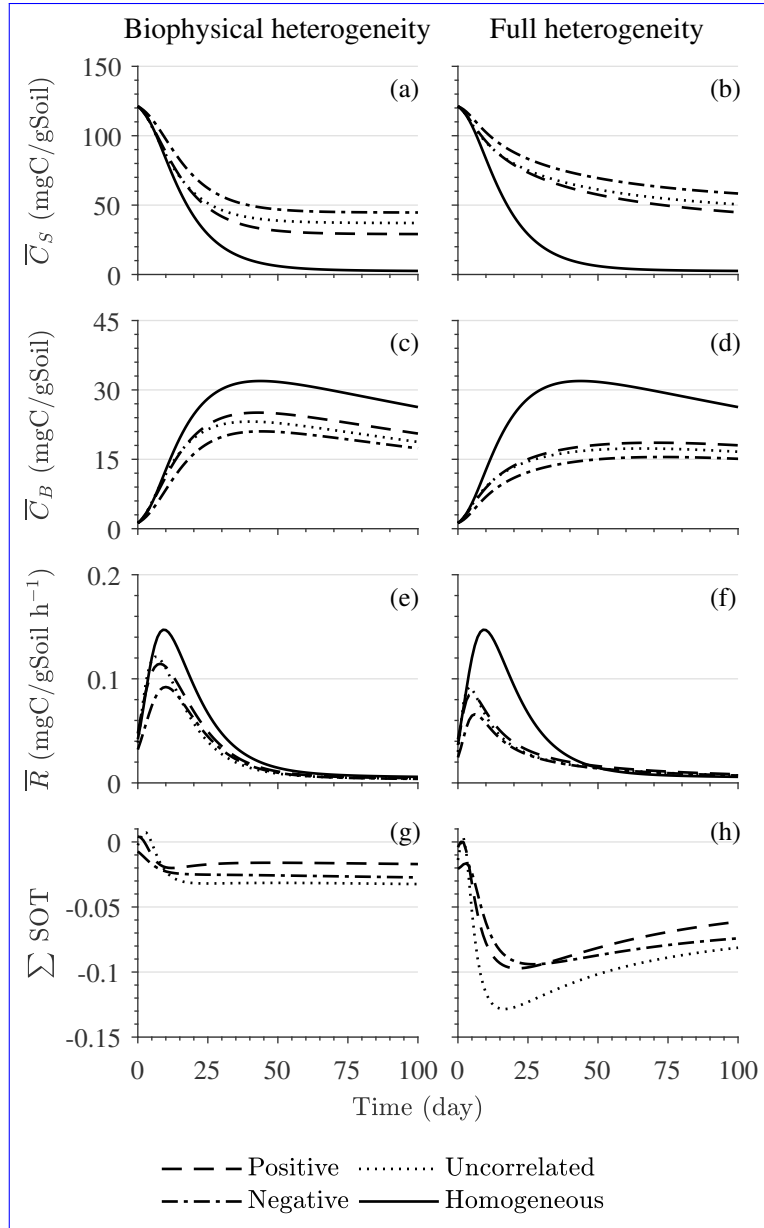


Figure 7. Temporal evolution of mean respiration rate in the heterogeneous system Scenario 2 (\bar{R}_{net} HS with inverse Michaelis-Menten kinetics), which includes the mean-field approximation: effect of biophysical heterogeneity (MFA left panel) and second-order terms, and full heterogeneity (right panel) on the respiration rate in macroscopic decomposition dynamics when the substrate is distributed around a value higher than the steady state of the homogeneous system: (\bar{R}_{nom}) a, for b) mean substrate C (a-e \bar{C}_s) biophysically and, (d-f, d) fully heterogeneous system with positive mean microbial C (a-b \bar{C}_b), negative (e-de, f) and un-correlated mean respiration rate (e-f \bar{R}) substrate, and microbial C for multiplicative kinetics (g, h) sum of second order terms ($\sum HOT$).

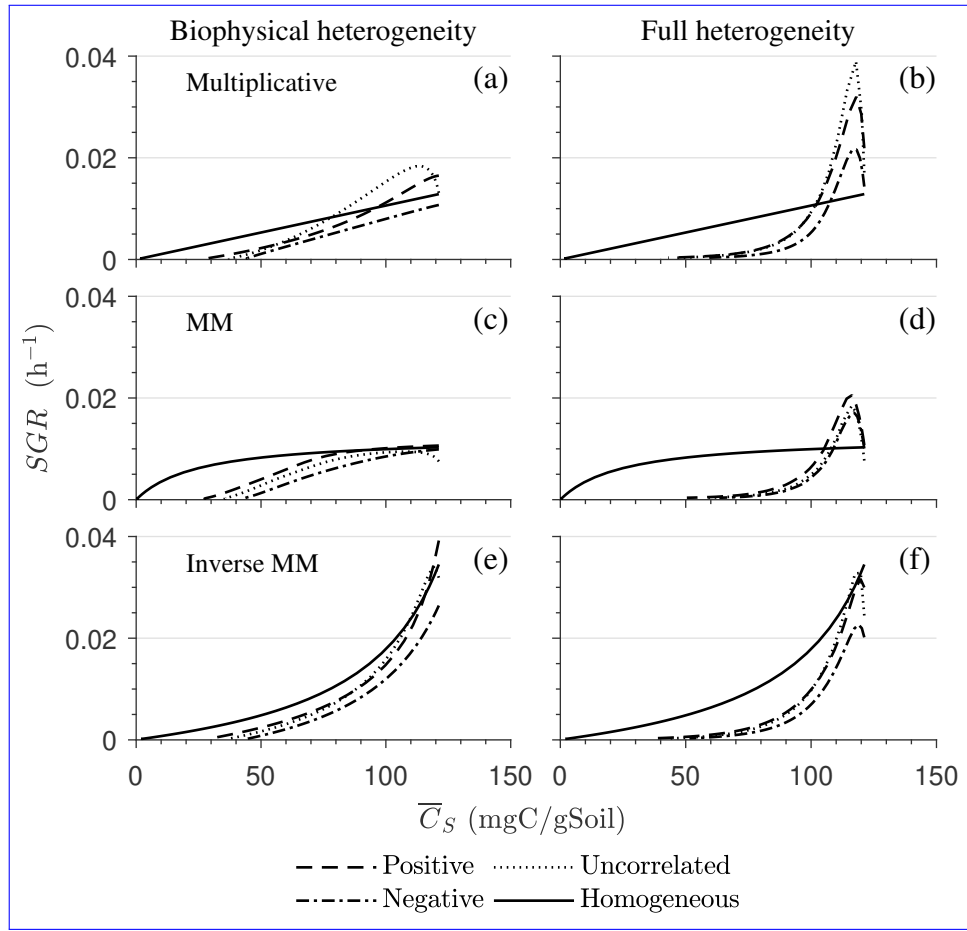


Figure 8. Effect of spatial heterogeneity on the mean specific growth rate (SGR) for HS scenario with the simplified micro-scale model (no C redistribution): Effect of biophysical (left column) and full (right column) heterogeneity on the mean specific growth rate (SGR) as a function of mean substrate C (\bar{C}_s) for the heterogeneous system for (a,b) multiplicative and, (c,d) Michaelis-Menten, and (e,f) inverse Michaelis-Menten kinetics. Time progresses from right to left, as \bar{C}_s is depleted.

the macro-scale equations that depend on the spatial distribution of organic matter and microorganisms (i.e., the second order spatial moments, SOT).

These second order moments capture fluxes that occur at the macro-scale as a result of nonlinear interactions in heterogeneous environments, but that do not occur in a homogeneous environment. terms represent corrections to the mean-field approximation

5 and depend on the spatial variability and co-variation of the state variables (i.e. C_s and C_b). Our numerical results showed that the second order spatial moments have their own dynamics that drive the heterogeneous system away from the mean-field approximation. Notably, while it is recognized that spatial distributions at the micro-scale affect macro-scale dynamics (Fal-

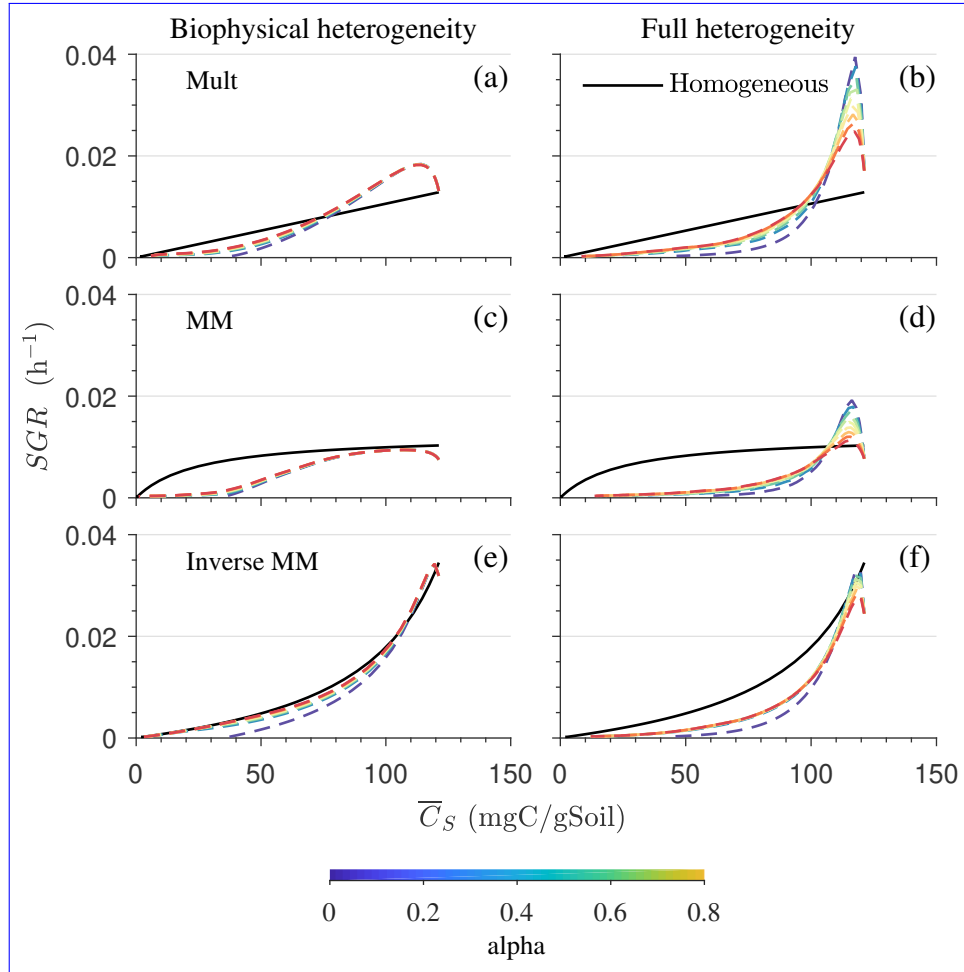


Figure 9. Effect of spatial heterogeneity on the mean specific growth rate (SGR) for HS scenario including C redistribution : Effect of biophysical (left column) and full (right column) heterogeneity on the mean SGR as a function of mean substrate C (\bar{C}_s) for an uncorrelated initial distribution of substrates and microorganisms. The three 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 \bar{C}_s is depleted.

coner et al., 2015), none of the current spatially-lumped SOM models include second or higher order terms that depend on micro-scale heterogeneity (see Sect. 4.3).

The simplicity of the micro-scale model and the derived analytical expressions are such that specific insights on how heterogeneity shapes micro-scale decomposition patterns can be gleaned and hypotheses generated. The ~~four~~ main predictions of this model are

1. Perturbing a system initially homogeneous and at steady state by redistributing substrates triggers fluctuations around the steady state (Fig. 4).
2. When only biophysical heterogeneity occurs, in the early microbial growth phase, macroscopic C fluxes are enhanced by co-location of substrates and microorganisms, and suppressed when they are isolated (Fig. 8a–c).
3. Combined biophysical and biochemical heterogeneity enhance C fluxes in the early stage of decomposition and suppress it in the later stages, compared to a homogeneous system (Fig. 8b–d).
4. Both biophysically and fully heterogeneous systems result in a transient persistence of SOM (Fig. 5a,d and 6a,d). ~~This persistence vanishes for~~ In the biophysically heterogeneous ~~in steady state (i.e. all the carbon is decomposed provided enough time; Fig. 4); however,~~ system at steady state all C is eventually decomposed, whereas in the fully heterogeneous system ~~retains this persistence even at steady state (i.e. not all C is decomposed~~ more C is retained as the substrate pool reaches a new equilibrium (Appendix A3).
5. For a successive reduction in the degree of heterogeneity (i.e. systematically moving from a heterogeneous to a homogeneous system), macro-scale dynamics converge to the mean-field approximation; i.e., the same kinetics can be used at all scales (Sect. 2.1.2).
6. Increasing local connectivity among grid cells moderately reduces the effect of spatial heterogeneity on the macro-scale variables and fluxes.
7. The inverse Michaelis-Menten kinetics appear to be less sensitive to the scale transition than multiplicative and Michaelis-Menten kinetics, but this result might depend on the specific choice of parameter values (for a discussion on scale invariance of upscaled kinetics for reaction networks, see Tang and Riley, 2017).
- 25 Our analysis suggests that the persistence of SOM in heterogeneous systems may be a consequence of the micro-scale heterogeneity in soil carbon cycling. In the transient simulations with biophysical heterogeneity, persistence is a result of

spatial disconnection between substrate and microorganism, captured in our framework by a low probability of co-location at the beginning of the simulation. In the transient simulations for the fully heterogeneous systems, persistence is a result of the combined effects of low probability of co-location and high probability of low decomposition rate constant at the beginning of the simulation. The heterogeneity in substrate quality thus explains the higher persistence of SOM in the fully heterogeneous system compared to the biophysically heterogeneous system.

4.2 Linking theory and observations

We studied three initial heterogeneous distributions of substrate and microbial C; positive, negative or no correlation between these two variables. These heterogeneities may correspond to spatial aggregation, isolation or random occurrence of substrate and microorganisms, respectively. Spatial aggregation is expected in litter and in the surface soil where substrate is abundant and microbial colonies are formed around hot spots (Nunan et al., 2003). Spatial isolation is more likely to occur in the subsoil because of lower substrate and microorganism density as well as poor pore connectivity (Ekschmitt et al., 2008; Salomé et al., 2010), and C-rich patches occur around roots that are separated by large (in a relative sense) volumes of soil that only receive diluted resources via percolation, diffusion, and bioturbation (Kuzyakov and Blagodatskaya, 2015). Uncorrelated spatial fields of substrate and microorganisms may correspond to spatial distributions between these two extremes. There are other examples of contrasting homogeneous vs. heterogeneous conditions. Disturbed, sieved or dispersed samples ~~can~~may be considered as homogeneous, whereas intact soil samples retain their natural heterogeneity.

Despite the correspondence of our idealized heterogeneity scenarios with ~~actual~~ conditions in natural soils or soil samples, linking our model predictions to observations is challenging, mostly because the effects of heterogeneity can not be easily isolated in experiments or field observations.

~~Many studies have focused on understanding~~ For example, experiments studying the effects of soil structure on the dynamics of SOM mineralization ~~, either by changing the pore network by controlling the water potential (Killham et al., 1993; Ruamps et al., 2011), or by artificially altering the soil structure through mechanical treatments (Stenger et al., 2002; Juarez et al., 2013; Negassa et al., 2015; Herbst et al., 2016). Changes in soil structure affect spatial heterogeneity of substrate and microbial C (Ruamps et al., 2011; Sleutel et al., 2012; Vos et al., 2013), but also~~ (Killham et al., 1993; Stenger et al., 2002; Ruamps et al., 2011; Juarez et al., 2013; Negassa et al., 2015; Herbst et al., 2016)

may introduce other types of heterogeneities that are not dealt with here. For example, samples with different pore networks (Ruamps et al., 2011) likely exhibit different water and air diffusive pathways, which in turn affect microbial respiration ~~(Moyano et al., 2013; Manzoni and Katul, 2014; Herbst et al., 2016; Koestel and Schlüter, 2019). Furthermore, the same spatial distribution of decomposers and substrates in different pore sizes may result in different C fluxes because the size of the~~

pores where decomposers reside affects the rate at which substrate can become available. Therefore, these studies may be ideal for studying the effect of soil structure, but not for isolating the spatial heterogeneity effects. (Manzoni and Katul, 2014; Herbst et al., 2016;

When targeting experiments to test our up-scaled equations, using data from previous studies is challenging because (i) spatial distribution of substrate and/or microbial C are not changed in a controlled manner and (ii) the scale at which physical treatments are performed is probably larger than the scale at which heterogeneity affects C dynamics.

We also found that SOT are highly dynamic, suggesting that the role of heterogeneities varies through time. However, in most long-term incubation experiments measurement frequency is low. Fraser et al. (2016) observed significant differences in the respiration dynamics using a very high frequency (6 minutes interval) measurements that is generally lost in daily or sub daily observations. In a recent review Baveye et al. (2018) suggested to improve the temporal resolution of observations of the soil physical environment. Extending this idea to biochemical processes is important because a complete description of macro-scale C fluxes requires spatial and dynamic measurements of the second order moments (see Sect. 4.4).

Thus, experiments in which soil structure was manipulated do not allow direct testing of the predicted links between heterogeneity and decomposition kinetics at the macro-scale. Therefore, while the Furthermore, an experimental validation of the present work could stem from designing a microscale experiments using artificial porous media with different degrees of heterogeneity. Recent application of microfluidics in soil science (Stanley et al., 2016; Aleklett et al., 2018) could allow isolating the effect of spatial heterogeneity. If any difference is observed among heterogeneous systems, then our framework could be used to attribute these differences to spatial heterogeneity at the micro-scale. While the proposed mathematical framework is conceptually useful, it is thus challenging to test. Nevertheless, the prediction that co-location of microorganisms and substrates promotes decomposition is consistent with and explains theoretically the results of recent experiments (Don et al., 2013; Schnecko

4.3 Developing soil carbon cycling models that account for micro-scale heterogeneity

Historically, the linear microbial implicit models were developed to explain long-term loss of C from agricultural soils or regional-scale variations in SOM (Jenny et al., 1949; Olson, 1963; Jenkinson and Rayner, 1977; Parton et al., 1987). However, when applied at fine spatial or temporal scale, these models fail to describe the dynamics of SOM (Zelenev et al., 2000; Manzoni and Porporato, 2007). To fill this gap and describe microbial processes at the macro-scale, nonlinear microbial explicit models have been proposed (Schimel and Weintraub, 2003; Manzoni and Porporato, 2009; Xie, 2013). In contrast to these approaches that impose nonlinear kinetics at the macro-scale, here we started from the assumption that SOM kinetics are either linear or nonlinear at the micro-scale, and let scale transition theory determine the type of kinetics at the macro-scale.

Conceptually, this approach is similar to upscaling chemical reaction networks to obtain a compact kinetic law that only depends on the concentrations of reactants and products (Tang and Riley, 2013, 2017). However, here we focus on spatial heterogeneity rather than on the complexity of chemical reactions. In a more complete upscaling approach, both sources of micro-scale variability should be taken into account.

5 When assuming linear kinetics at the micro-scale, we showed analytically that the kinetics at the macro-scale remain linear and independent of soil biophysical heterogeneity (Eq. (17) and (22)). This result has implications for experimental studies linking soil architecture to SOM mineralization. In some of these studies, first order microbial implicit kinetics are used to describe the data (Bouckaert et al., 2013; Juarez et al., 2013). ~~This means that if~~ If a linear model captures well the SOM dynamics in a heterogeneous system, then either the underlying micro-scale dynamics are indeed linear, or the averaging of
10 underlying nonlinear equations leads to linearity at macro-scale.

Conversely, we demonstrate that nonlinear kinetics at micro-scale do not remain the same when scaling up. The macro-scale dynamics retain a clear signature of nonlinearities at the micro-scale in the MFA term, but the second order terms could be even more important than the MFA. Thus, nonlinear kinetics might improve SOM predictions because microbial activity is accounted for (Wieder et al., 2013) (at the cost of increased uncertainty, Wieder et al. (2018)), but the question remains: which
15 ~~kinetics~~ kinetic formulation should be used at the macro-scale that captures both microbial activity and spatial heterogeneities? We offer a framework to advance this area by using appropriately upscaled nonlinear kinetics including SOT at macro-scale. This upscaling framework can be extended to account for the role of other micro-scale interactions such as among substrates, microorganisms, and minerals, or even temporally varying connectivity due to water movement. These improvements, however, would come at the expense of increased number of nonlinear second order spatial moments.

20 To summarize, the proposed theoretical developments allow integration of spatial heterogeneity into decomposition kinetics. Assuming that the second order spatial moments are known, this integration can be achieved by using the equations listed in Table 2 instead of standard linear or nonlinear ~~kinetics~~ kinetic equations used in the current models (Wieder et al., 2018; Abramoff et al., 2018). However, the second order moments and their dynamics are not known in general, as discussed at the end of the following section.

25 4.4 Limitations of the upscaling approach

To illustrate the effects of spatial heterogeneities alone, we simulated idealized laboratory conditions in which the environmental conditions are constant so that the decomposition rate is not affected by soil moisture and temperature changes through time and space. Moreover, ~~we did not account for the spatial redistribution of organic matter and microbial biomass caused by~~

diffusion/dispersion/advection or bioturbation in the distributed model. Finally, the simulated domain is small compared to an actual soil sample, but we regard the number of simulated grid cells (10^4) as representative of the range of variation occurring in larger, similarly idealized samples. In other studies, more complex micro-scale models based on nonlinear reactive and diffusive fluxes have been implemented (Monga et al., 2008; Nguyen-Ngoc et al., 2013; Monga et al., 2014); however, their spatial upscaling would require volume averaging of the coupled transport and reaction equations, making the problem mathematically intractable when aiming for analytical solutions (Whitaker, 1999; Valdés-Parada et al., 2009; Porter et al., 2011; Lugo-Méndez et al., 2015). The two pool micro-scale model with initial heterogeneous distributions of substrate and microorganisms as described in this study offers a simplified way of simulating reaction-diffusion systems. The two end-member cases of homogeneous and fully heterogeneous systems where grid cells are independent are representative of conditions in which diffusivities are high compared to reaction kinetics in the former and negligible in the latter. In more realistic settings, conditions are likely to be intermediate between these two cases, as described by varying the value of the mass transfer coefficient α (Fig. 9).

Including C redistribution as a simple mass transfer process does not allow studying how soil structure affects macro-scale dynamics by creating and maintaining heterogeneous distributions of resources and oxygen, such as in soil aggregates (Keiluweit et al., 2011). These patterns result from the interaction of transport and reaction processes that the proposed idealized models cannot capture.

The upscaling mechanism described in this work assumes that microbial mortality is first order in microbial C, so that this term remains structurally similar in the macroscopic Eq. (13) and (14). A nonlinear mortality generalized by $T = k_B C_b^\beta$ (Georgiou et al., 2017) would create an additional term in the macro-scale equations. The mean microbial mortality can be calculated by inserting the nonlinear T into Eq. (16), resulting in $\bar{T} = k_B \bar{C}_b^\beta + \beta \bar{C}_b^{\beta-1} \sigma_{C_s}^2$, where $\sigma_{C_s}^2$ is the spatial variance of microbial C (for the biophysically heterogeneous system; i.e., k_B is spatially invariant). For $\beta = 1$, the first order mortality is recovered, (Eq. (22)); for $\beta \neq 1$, \bar{T} has an additional positive variance term that increases mortality at the macro-scale.

Finally, the upscaled macro-scale equations still require a closure scheme for integration; i.e., a set of equations linking the spatial moments to the mean state variables. With such a set of additional equations, the problem becomes mathematically 'closed', as the only remaining unknowns are the mean state variables. Examples of closure from other fields are mentioned in the introduction (e.g., Bergström et al. (2006)), but finding a robust closure scheme remains challenging and will be the subject of future work.

Moreover, our derivations are general, but how these closure equations are formulated and parameterized will likely depend on the scale transition under consideration - soil pore to core (as in this work), soil core to field, or even field to landscape. It

is possible that a whole hierarchy of scale transitions is required to determine macro-scale equations suitable for regional or global-scale applications. Along similar lines, how many terms in the Taylor expansion should be retained at each level of this hierarchy remains an open question. It is also possible that the dynamics at the micro scale in combination with C redistribution lead to low values of higher order moments, thus allowing us to neglect higher order terms– because substrate consumption, mortality of the microorganisms, and transport contribute to smoothing spatial gradients.

5 Conclusions and perspective

Most carbon cycling models implicitly assume a spatially homogeneous distribution of SOM in different C pools and are based on the mean-field approximation of the rate of decomposition. However, assuming homogeneity is adequate only at the micro-scale in soils, due to the homogenizing effect of diffusion, which brings carbon sources and decomposers into direct contact with each other at such scales. Therefore, the mean-field approximation is valid only at the micro-scale, creating a challenge when developing SOM models at macro-scale that also account for environment heterogeneity. In this contribution, we used scale transition theory to link an idealized (but realistic) heterogeneous system and a homogeneous system by establishing an analytical expression for the macroscopic mean decomposition rate that accounts for the micro-scale heterogeneities. Unlike the mean-field approximation adopted in most C cycling models, the upscaled governing equations we derived include second order spatial moments; i.e., spatial variances and/or covariances between micro-scale state variable and model parameters. The dynamical behavior of the second order terms drives the heterogeneous system away from the mean-field approximation. For a heterogeneous system, initially near steady state, micro-scale heterogeneities led to oscillations in the macro-scale respiration flux and higher SOM persistence in a fully heterogeneous system. For a heterogeneous system perturbed from its equilibrium, the co-location of substrate and microorganisms increased macroscopic C fluxes compared to a case in which they were isolated.

In conclusion, this work provides a methodology to explicitly include micro-scale heterogeneity in soil C cycling models. Our upscaled kinetic equations could be used in lieu of current formulations, but additional equations describing the dynamics of spatial moments should be further developed to mathematically close the problem. ~~These~~ The upscaled equations show that, (i) heterogeneities alter the form of the carbon flux equations at the macro-scale and, as a result, (ii) co-location (respectively isolation) of microorganisms and their substrates promote (suppress) carbon fluxes in soils.

Code availability. The codes used to construct the heterogeneous soil maps and to solve the mass balance equations in heterogeneous domains are publicly available via DOI <https://doi.org/10.5281/zenodo.3253880>.

Data availability. The article does not use any relevant data.

Appendix A

5 A1 Derivation of the macro-scale rate of decomposition

Here we describe the derivation of the spatially averaged C flux for a generic microscopic C flux $F(C_s, C_b, k)$ using scale transition theory. As a first step, we calculate the multi-variate Taylor's series expansion of $F(C_s, C_b, k)$ around the spatial average value of C_s , C_b and k ,

$$\begin{aligned}
 F(C_s, C_b, \mathbf{k}) &= F(\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}) + \frac{\partial F}{\partial C_s} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_s - \bar{C}_s) + \frac{\partial F}{\partial C_b} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_b - \bar{C}_b) + \frac{\partial F}{\partial C_b} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_b - \bar{C}_b) + \\
 10 \quad &\sum_{i=1}^n \frac{\partial F}{\partial k_i} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (k_i - \bar{k}_i) + \frac{1}{2} \frac{\partial^2 F}{\partial C_s^2} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_s - \bar{C}_s)^2 + \frac{1}{2} \frac{\partial^2 F}{\partial C_b^2} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_b - \bar{C}_b)^2 + \\
 &\frac{1}{2} \sum_{i=1}^n \sum_{j=1}^n \frac{\partial^2 F}{\partial k_i \partial k_j} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (k_i - \bar{k}_i)(k_j - \bar{k}_j) + \frac{\partial^2 F}{\partial C_s \partial C_b} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_s - \bar{C}_s)(C_b - \bar{C}_b) + \\
 &\sum_{i=1}^n \frac{\partial^2 F}{\partial k_i \partial C_s} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_s - \bar{C}_s)(k_i - \bar{k}_i) + \sum_{i=1}^n \frac{\partial^2 F}{\partial k_i \partial C_b} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_b - \bar{C}_b)(k_i - \bar{k}_i) + O(C_s^3 C_b^3 k_i^3), \quad (A1)
 \end{aligned}$$

where $O(C_s^3, C_b^3, k_i^3)$ represents the higher order terms and the overbars denote the spatially averaged micro-scale quantities.

Second, the averaging operator given by Eq. (1) is applied in Eq. (A1). Truncating terms above the second order terms, Eq. (A1) becomes

$$\begin{aligned}
& \frac{1}{\int \int dxdy} \int \int F(C_s, C_b, \mathbf{k}) dxdy = \\
& \frac{1}{\int \int dxdy} \left[F(\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}) + \frac{\partial F}{\partial C_s} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_s - \bar{C}_s) + \frac{\partial F}{\partial C_b} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_b - \bar{C}_b) + \frac{\partial F}{\partial C_b} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_b - \bar{C}_b) + \right. \\
& \sum_{i=1}^n \frac{\partial F}{\partial k_i} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (k_i - \bar{k}_i) + \frac{1}{2} \frac{\partial^2 F}{\partial C_s^2} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_s - \bar{C}_s)^2 + \frac{1}{2} \frac{\partial^2 F}{\partial C_b^2} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_b - \bar{C}_b)^2 + \\
& \frac{1}{2} \sum_{i=1}^n \sum_{j=1}^n \frac{\partial^2 F}{\partial k_i \partial k_j} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (k_i - \bar{k}_i)(k_j - \bar{k}_j) + \frac{\partial^2 F}{\partial C_s \partial C_b} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_s - \bar{C}_s)(C_b - \bar{C}_b) + \\
& \left. \sum_{i=1}^n \frac{\partial^2 F}{\partial k_i \partial C_s} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_s - \bar{C}_s)(k_i - \bar{k}_i) + \sum_{i=1}^n \frac{\partial^2 F}{\partial k_i \partial C_b} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} (C_b - \bar{C}_b)(k_i - \bar{k}_i) \right]. \quad (\text{A2})
\end{aligned}$$

In Eq. (A2), the first order partial derivatives (second, third and fourth term between the square brackets) disappear, because the partial derivatives evaluated at the mean state variables are constants that are multiplied by the expectation of the deviation of a quantity, which is zero $\left(\frac{\partial F}{\partial \chi} \Big|_{\bar{\chi}} \int \int (\chi - \bar{\chi}) dxdy = 0 \right)$, where χ is C_s, C_b or k .

Finally, after applying the averaging operator, deviations multiplying the second order partial derivatives become spatial variances and covariances. As a result, the macro-scale C flux $\bar{F}(C_s, C_b, \mathbf{k})$ is obtained

$$\begin{aligned}
\bar{F}(C_s, C_b, \mathbf{k}) = & F(\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}) + \frac{1}{2} \frac{\partial^2 F}{\partial C_s^2} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} \sigma_{C_s}^2 + \frac{1}{2} \frac{\partial^2 F}{\partial C_b^2} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} \sigma_{C_b}^2 + \sum_{i=1}^n \sum_{j=1}^n \frac{\partial^2 F}{\partial k_i \partial k_j} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} \overline{k'_i k'_j} \\
& \frac{\partial^2 F}{\partial C_s \partial C_b} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} \overline{C'_s C'_b} + \sum_{i=1}^n \frac{\partial^2 F}{\partial k_i \partial C_s} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} \overline{k'_i C'_s} + \sum_{i=1}^n \frac{\partial^2 F}{\partial k_i \partial C_b} \Big|_{\bar{C}_s, \bar{C}_b, \bar{\mathbf{k}}} \overline{k'_i C'_b}, \quad (\text{A3})
\end{aligned}$$

Equation (A3) can be used to obtain the macro-scale C flux given the decomposition function D at micro-scale. Explicit solutions for the multiplicative kinetics are reported in the main text (Eq. (18), (20), and (21)).

For illustration, here we report the derivation of the spatially averaged rate of decomposition for Michaelis-Menten (MM) kinetics. The micro-scale rate of decomposition for MM kinetics is given by

$$F = \frac{k_{s,mm} C_s C_b}{K_M + C_s} \frac{k_{MM} C_s C_b}{K_{MM} + C_s} \quad (\text{A4})$$

where both the parameters $\underline{k_{s,mm}}$ and $\underline{K_M}$ and $\underline{k_{MM}}$ and $\underline{K_{MM}}$ and the state variables C_s and C_b are considered spatially variable quantities. Inserting Eq. (A4) into Eq. (16) gives the macro-scale rate of decomposition

$$\begin{aligned} \bar{F}(C_s, C_b, [k_{s,mm}, K_M, k_{MM}, K_{MM}]) = & F(\bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM}) + \frac{1}{2} \frac{\partial^2 F}{\partial C_s^2} \left| \bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM} \right| \sigma_{C_s}^2 + \frac{1}{2} \frac{\partial^2 F}{\partial k_{s,mm}^2} \frac{\partial^2 F}{\partial k_{MM}^2} \left| \bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM} \right| \sigma_{k_{s,mm}}^2 \\ & + \frac{1}{2} \frac{\partial^2 F}{\partial k_{MM}^2} \left| \bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM} \right| \sigma_{k_{MM}}^2 + \frac{\partial^2 F}{\partial k_{s,mm} \partial k_{MM}} \frac{\partial^2 F}{\partial k_{MM} \partial K_{MM}} \left| \bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM} \right| \sigma_{k_{s,mm}} \sigma_{k_{MM}} \\ & + \frac{\partial^2 F}{\partial C_s \partial C_b} \left| \bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM} \right| \bar{C}_s' \bar{C}_b' + \frac{\partial^2 F}{\partial k_{s,mm} \partial C_s} \frac{\partial^2 F}{\partial k_{MM} \partial C_s} \left| \bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM} \right| \bar{C}_s' \bar{C}_b' \\ & + \frac{\partial^2 F}{\partial k_{s,mm} \partial C_b} \frac{\partial^2 F}{\partial k_{MM} \partial C_b} \left| \bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM} \right| \bar{C}_b' \bar{C}_b' + \frac{\partial^2 F}{\partial K_M \partial C_s} \frac{\partial^2 F}{\partial K_{MM} \partial C_s} \left| \bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM} \right| \bar{C}_s' \bar{K}_M' \\ & + \frac{\partial^2 F}{\partial K_M \partial C_b} \frac{\partial^2 F}{\partial K_{MM} \partial C_b} \left| \bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM} \right| \bar{C}_b' \bar{K}_{MM}'. \end{aligned} \quad (A5)$$

The partial derivative of F with respect to $\underline{k_{s,mm}}$ and $\underline{k_{MM}}$ is zero because F is a linear function of $\underline{k_{s,mm}}$ and $\underline{k_{MM}}$. Now, for biophysical heterogeneous and biochemical homogeneous system, covariances and variances related to parameters are zeros so that we are left with

$$\bar{F}(C_s, C_b, [k_{s,mm}, K_M, k_{MM}, K_{MM}]) = F(\bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM}) + \frac{1}{2} \frac{\partial^2 F}{\partial C_s^2} \left| \bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM} \right| \sigma_{C_s}^2 + \frac{\partial^2 F}{\partial C_s \partial C_b} \left| \bar{C}_s, \bar{C}_b, \bar{k}_{s,mm}, \bar{K}_M, \bar{k}_{MM}, \bar{K}_{MM} \right| \sigma_{C_s} \sigma_{C_b} \quad (A6)$$

Calculating the derivatives gives

$$\bar{F}(C_s, C_b, [k_{s,mm}, K_M, k_{MM}, K_{MM}]) = \frac{k_{s,mm} \bar{C}_s \bar{C}_b}{K_M + \bar{C}_s} \frac{k_{MM} \bar{C}_s \bar{C}_b}{K_{MM} + \bar{C}_s} + \frac{1}{2} \left[\frac{-2k_{s,mm} K_M \bar{C}_b}{(K_M + \bar{C}_s)^2} \frac{-2k_{MM} K_{MM} \bar{C}_b}{(K_{MM} + \bar{C}_s)^3} \right] \sigma_{C_s}^2 + \left[\frac{k_{s,mm} K_M}{(\bar{C}_s + K_M)^2} \frac{k_{MM} K_{MM}}{(\bar{C}_s + K_{MM})^2} \right] \sigma_{C_s} \sigma_{C_b} \quad (A7)$$

For biophysical homogeneous and biochemical heterogeneous system, covariances and variances of state variables (C_s and C_b) are zeros so that we are left with

$$\begin{aligned}
& \overline{F}(C_s, C_b, \left[\underline{k_{s,mm}MM}, \underline{K_{MM}} \right]) = \\
& F(\overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M) + \frac{1}{2} \frac{\partial^2 F}{\partial k_M^2} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{MM}MM}, \overline{K}_M \sigma_{k_M}^2 + \\
& \frac{\partial^2 F}{\partial k_{s,mm} \partial k_M} \frac{\partial^2 F}{\partial k_{MM} \partial K_{MM}} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{MM}MM}, \overline{K}_M \overline{k'_{MM} K'_{MM}} + \frac{\partial^2 F}{\partial k_{s,mm} \partial C_s} \frac{\partial^2 F}{\partial k_{MM} \partial C_s} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{MM}MM}, \overline{K}_M \overline{C'_s C'_b} \\
& \frac{\partial^2 F}{\partial k_{s,mm} \partial C_b} \frac{\partial^2 F}{\partial k_{MM} \partial C_b} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{MM}MM}, \overline{K}_M \overline{C'_b K'_{MM}} + \frac{\partial^2 F}{\partial K_M \partial C_s} \frac{\partial^2 F}{\partial K_{MM} \partial C_s} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{MM}MM}, \overline{K}_M \overline{C'_b K'_{MM}}. \tag{A8}
\end{aligned}$$

For a completely heterogeneous system with biophysical and biochemical heterogeneity, the mean rate of decomposition at macro-scale is given by Eq. (A9).

$$\begin{aligned}
& \overline{F}(C_s, C_b, \left[\underline{k_{s,mm}MM}, \underline{K_{MM}} \right]) = \\
& F(\overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M) + \frac{1}{2} \frac{\partial^2 F}{\partial C_s^2} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \sigma_{C_s}^2 + \\
& \frac{1}{2} \frac{\partial^2 F}{\partial k_M^2} \frac{\partial^2 F}{\partial K_M^2} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \sigma_{k_M}^2 + \frac{\partial^2 F}{\partial k_{s,mm} \partial k_M} \frac{\partial^2 F}{\partial k_{MM} \partial K_{MM}} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \overline{k'_{MM} K'_{MM}} \\
& \frac{\partial^2 F}{\partial C_s \partial C_b} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \overline{C'_s C'_b} + \frac{\partial^2 F}{\partial k_{s,mm} \partial C_s} \frac{\partial^2 F}{\partial k_{MM} \partial C_s} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \overline{C'_s C'_b} \\
& \frac{\partial^2 F}{\partial k_{s,mm} \partial C_b} \frac{\partial^2 F}{\partial k_{MM} \partial C_b} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \overline{C'_b K'_{MM}} + \frac{\partial^2 F}{\partial K_M \partial C_s} \frac{\partial^2 F}{\partial K_{MM} \partial C_s} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \overline{C'_b K'_{MM}} \\
& \frac{\partial^2 F}{\partial K_M \partial C_b} \frac{\partial^2 F}{\partial K_{MM} \partial C_b} \left| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \right| \overline{C}_s, \overline{C}_b, \overline{k_{s,mm}MM}, \overline{K}_M \overline{C'_b K'_{MM}}. \tag{A9}
\end{aligned}$$

Similar to MM kinetics, the mean rate of decomposition for IMM kinetics can also be calculated as,

$$\begin{aligned}
& \overline{F}(C_s, C_b, [k_{IMM}, K_{IMM}]) = \\
& \frac{F(\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}) + \frac{1}{2} \frac{\partial^2 F}{\partial C_s^2} \Big|_{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \sigma_{C_b}^2 +}{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \\
& \frac{\frac{1}{2} \frac{\partial^2 F}{\partial K_{IMM}^2} \Big|_{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \sigma_{K_{IMM}}^2 + \frac{\partial^2 F}{\partial k_{IMM} \partial K_{IMM}} \Big|_{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \overline{k'_{IMM} K'_{IMM}} +}{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \\
& \frac{\frac{\partial^2 F}{\partial C_s \partial C_b} \Big|_{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \overline{C'_s C'_b} + \frac{\partial^2 F}{\partial k_{IMM} \partial C_s} \Big|_{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \overline{C'_s k'_{IMM}} +}{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \\
& \frac{\frac{\partial^2 F}{\partial k_{IMM} \partial C_b} \Big|_{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \overline{C'_b k'_{IMM}} + \frac{\partial^2 F}{\partial K_{IMM} \partial C_s} \Big|_{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \overline{C'_s K'_{IMM}} +}{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \\
& \frac{\frac{\partial^2 F}{\partial K_{IMM} \partial C_b} \Big|_{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \overline{C'_b K'_{IMM}}.}{\overline{C}_s, \overline{C}_b, \overline{k}_{IMM}, \overline{K}_{IMM}} \tag{A10}
\end{aligned}$$

A2 An analytical expression Initial 2D random fields of the substrate C at steady state for the multiplicative kinetics in the fully heterogeneous system The substrate and microbial Cat

The heterogeneous field of microbial C was created using a random field generator that provides 100×100 spatially correlated random numbers between -1 and 1 (Lennon, 2000). These values were then re-scaled by an appropriate mean and standard deviation of microbial C. To simulate the dead zones in the heterogeneous system, some grid cells were forced to have no microbial C (the obtained field is denoted $y_{i,j}$). Moreover, to allow comparison among simulations, the microbial C field was re-normalized to have a specified value of total initial microbial C,

$$C_{b,i,j} = \frac{y_{i,j}}{\sum_{i,j} y_{i,j}} C_{b,total} \text{ (fg)} \tag{A11}$$

It is assumed that the $C_{b,total}$ is equal to 1% of the total amount of substrate in the domain (Witter, 1996), which is in turn calculated as $C_{s,total} = \overline{C}_{s,0} \times N_x \times N_y$ where $\overline{C}_{s,0}$ is the initial mean substrate C in a single grid cell (fg). The amount of substrate C in any grid cell is limited by the maximum amount of C that the cell can accommodate, according to the density of organic matter (ρ_{SOM}), and assuming that 50% of organic matter on a mass basis is composed of organic C. The maximum

amount of substrate C that one cell can contain is thus given by

$$C_{max} = 0.5 \rho_{SOM} \text{ cell}_{volume} \text{ (fg)} \quad (\text{A12})$$

where cell_{volume} is the volume of a grid cell. The value of $\bar{C}_{s,0}$ was chosen so that the maximum C amount at a micro-site does not exceed C_{max} . To summarize, the obtained spatially heterogeneous random fields of microbial C and substrate C satisfy the following constraints: i) the total amount of organic C is set, ii) the total amount of microbial C is 1% of total organic C, iii) the maximum amount of C in a cell is set (Eq. (A12)), and iv) some grid cells have no microbial biomass.

A3 Steady state substrate C for the multiplicative kinetics in the fully heterogeneous systems

The substrate C at steady state for multiplicative ~~and-MM~~ kinetics in a homogeneous ~~system are or~~ biophysically heterogeneous system is given in Table 1 and restated here for convenience. ~~These expressions are also valid for biophysically heterogeneous systems.~~

$$C_{s,mult M}^* = \frac{k_B}{Y k_{s,mult}} \frac{k_B}{Y k_M} \quad (\text{A13})$$

$$C_{s,mm MM}^* = \frac{K_M k_B}{Y k_{s,mm} - k_B} \frac{K_{MM} k_B}{Y k_{MM} - k_B} \quad (\text{A14})$$

where * represents the steady state. Eq. (A13) and (A14) show that the steady state substrate C depends only on the ~~kinetics parameters~~ kinetic parameters and microbial C-use efficiency. Thus, if the kinetic parameters are spatially variable (i.e., fully heterogeneous system) then $C_{s,mult}^*$ ~~and~~ $C_{s,mm}^*$ C_M^* and C_{MM}^* are also spatially variable and different from the steady state values ~~in of~~ biophysically heterogeneous and homogeneous systems. Knowing the probability distributions of the kinetic parameters, the mean steady state substrate C in the fully heterogeneous system can be calculated as the mean value of $C_{s,mult}^*$ ~~or~~ $C_{s,mm}^*$ C_M^* or C_{MM}^* .

The mean value of a generic function, $g(x)$ is given by $\overline{g(x)} = \int_{-\infty}^{\infty} g(x) f_X(x) dx$, where $f_X(x)$ is the probability density function of x . For the multiplicative kinetics and assumed a $\log Uniform(a, b)$ distribution for $k_{s,mult}$ k_M , the mean value of $C_{s,mult}^*$ C_M^* is given by,

$$\bar{C}_{s,mult M}^* = \int_a^b \frac{k_B}{Y k_{s,mult}} \frac{k_B}{Y k_M} f(k_{s,mult M}) dk_{s,mult M} \quad (\text{A15})$$

where $f(k_{s,mult})f(k_M)$ is the probability density function of $k_{s,mult}k_M$ and given by $f(k_{s,mult}) = \frac{1}{(b-a)k_{s,mult} \log_e 10}$ $f(k_M) = \frac{1}{(b-a)}$
Inserting the expression of $f(k_{s,mult})f(k_M)$ in Eq. (A15) gives,

$$\bar{C}_{s,mult}^* = \frac{k_B [10^{-a} - 10^{-b}]}{Y(b-a) \ln(10)} \quad (A16)$$

A4 Sensitivity of fluctuations to changes in k_M in scenario 1

- 5 We performed two sensitivity analyses in which we altered the kinetic constant parameter for the multiplicative decomposition model k_M : 1) decreasing k_M in the biophysical heterogeneity–positively correlated C_s and C_b (Fig. A1) and increasing the heterogeneity of k_M (by increasing its standard deviation) in the full heterogeneity case (Fig. A2). From Fig. A1, it is clear that decreasing the rate constant increases the amplitude and wavelength of the oscillations. As shown in Fig. A2, increasing the heterogeneity of the rate constant increases the amount of undecomposed substrate C compared to a lower degree of heterogeneity (Fig. 4). This pattern can be explained using the analytical expression of the steady state substrate C (see Eq. (A16) in Appendix A3). For the increased heterogeneity case shown in Fig. 4, we used values of a and b as listed in Table B3 for biochemical heterogeneity 1 and multiplicative kinetics, where a and b have the same meaning as in Eq. (A16). The analytical expression for the steady state, evaluated with these values of a and b, results in exactly the same steady state of substrate C as simulated by the distributed model (i.e., 15 mgC/gSoil).
- 10
- 15 These fluctuations are similar to those noted in earlier papers using spatially lumped models (Manzoni and Porporato, 2007; Sierra and M. . These papers showed that the occurrence and amplitude of the fluctuations depend on the kinetic parameter values, as is the case here.

Comparison of soil organic matter decomposition dynamics of linear, multiplicative and Michaelis-Menten kinetics. The rate constant of decomposition for multiplicative and linear kinetics was estimated by fitting the of the dynamic equations for the two models to the CO_2 production rate simulated by MM kinetics.

20

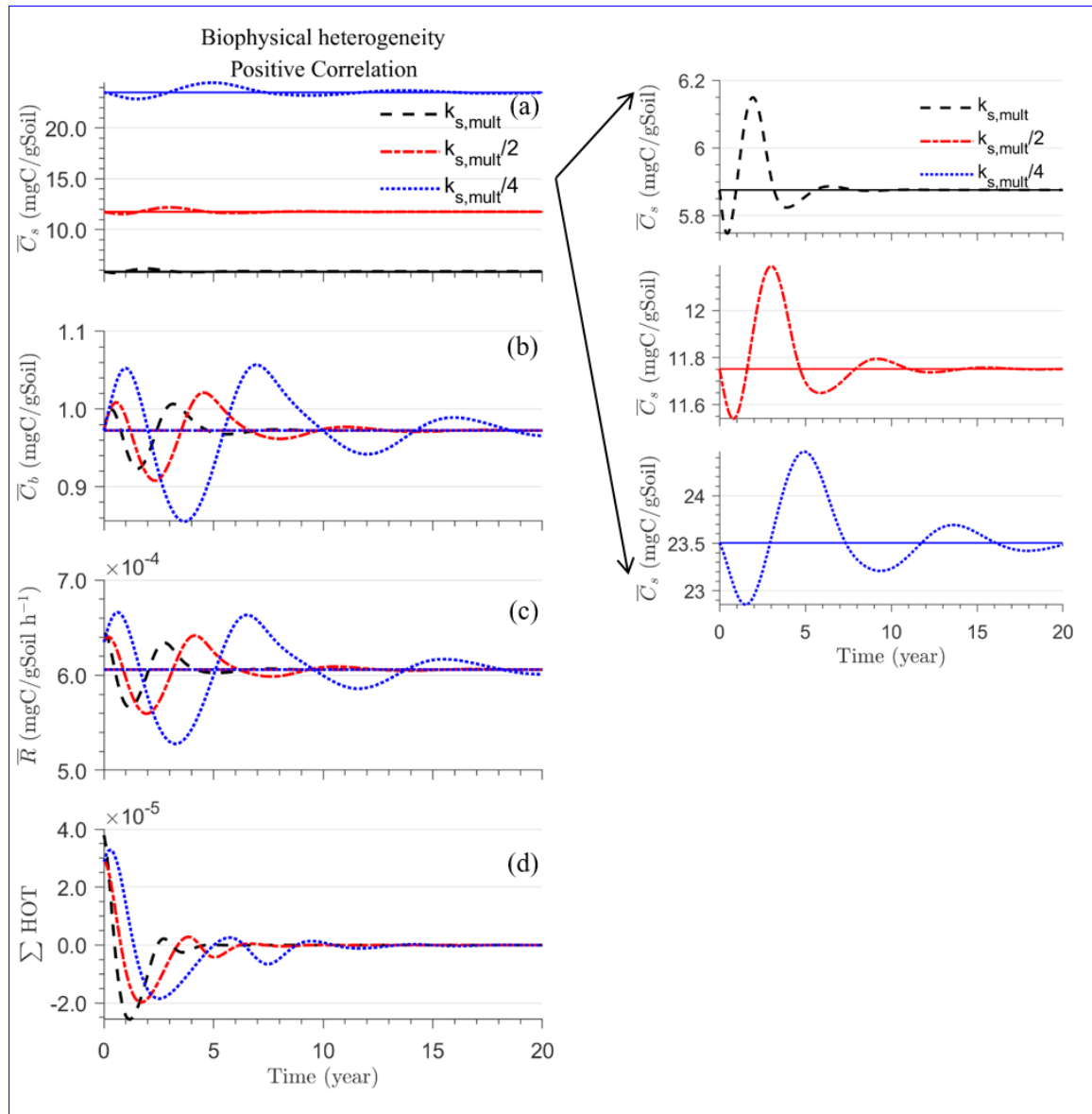


Figure A1. Steady-state-initial condition: Examples of the homogeneous (a) and the heterogeneous distributions of mean substrate C constrained to have the same amount (\bar{C}_s), (b) mean microbial C (\bar{C}_b), (c) mean respiration rate (\bar{R}), and (d) sum of total-substrate second and third order terms ($\sum HOT$) are shown as a function of time, for positively correlated initial spatial heterogeneity of C_s and C_b . The fields in b-d were obtained. This figure is similar to Fig. 4, left column (initial substrate is distributed randomly around the steady state). Varying levels of the rate constant k_M are shown (as indicated by imposing different degrees line styles and colors; the base case is the same as in Fig. 4). Panels on the right are enlarged views of correlation with the initial heterogeneous distributions time trajectories of microbial C (e) \bar{C}_s .

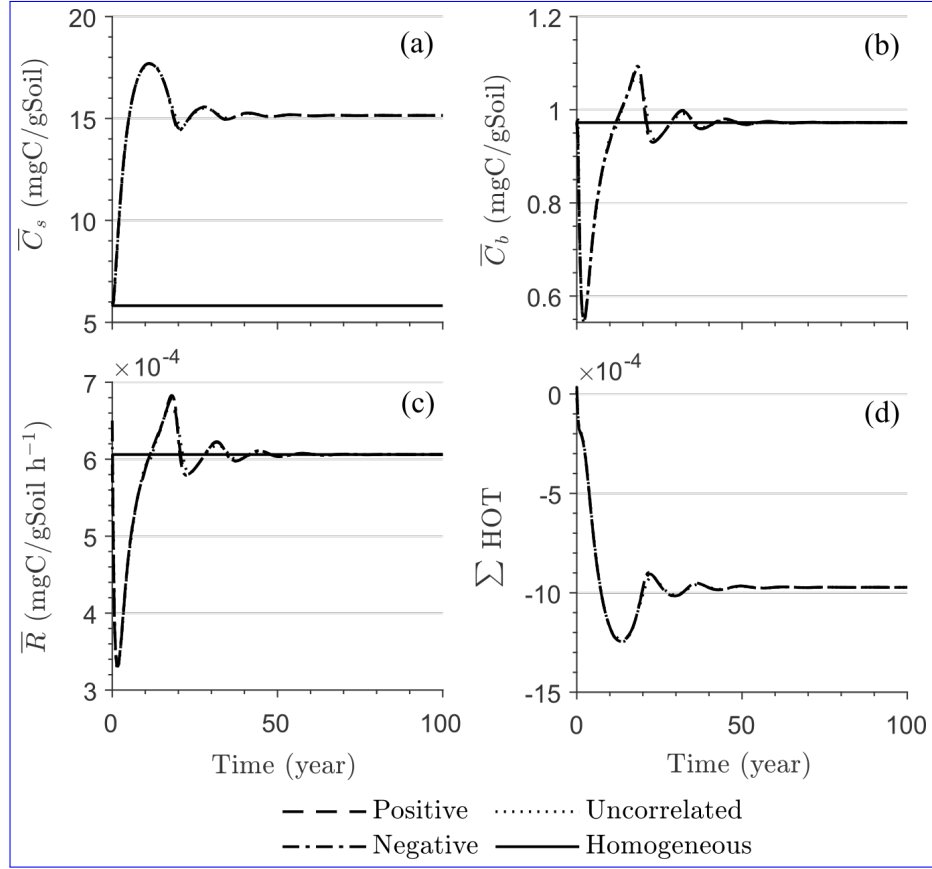


Figure A2. (a) mean substrate C (\bar{C}_s), (b) mean microbial C (\bar{C}_b), (c) mean respiration rate (\bar{R}), and (d) sum of second and third order terms (ΣHOT) are shown as a function of time, for different scenarios of initial spatial heterogeneity. This figure is similar to Fig. 4 for the full heterogeneity case, but with increased heterogeneity of the rate constant (k_M).

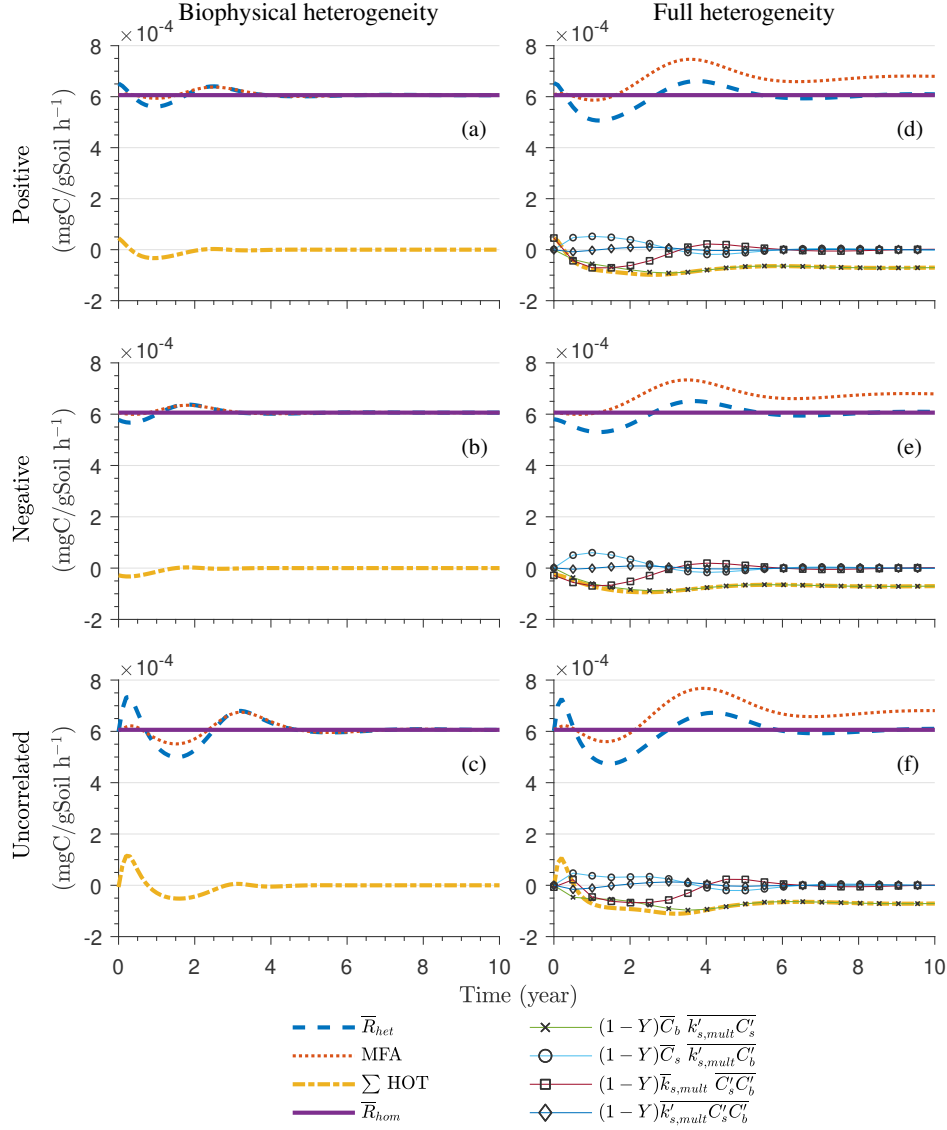


Figure A3. Scenario 1 (initial condition at steady state [simulation](#)): temporal evolution of mean respiration rate in the heterogeneous system (\bar{R}_{het} , including the mean-field approximation (MFA) and second order terms), and the respiration rate in the homogeneous system (\bar{R}_{hom}), for multiplicative kinetics and for (a–c) [the](#) biophysical and (d–f) [the](#) fully heterogeneous system with (a–d) positively and (b–e) negatively correlated, or (c–f) uncorrelated initial substrate and microbial C.

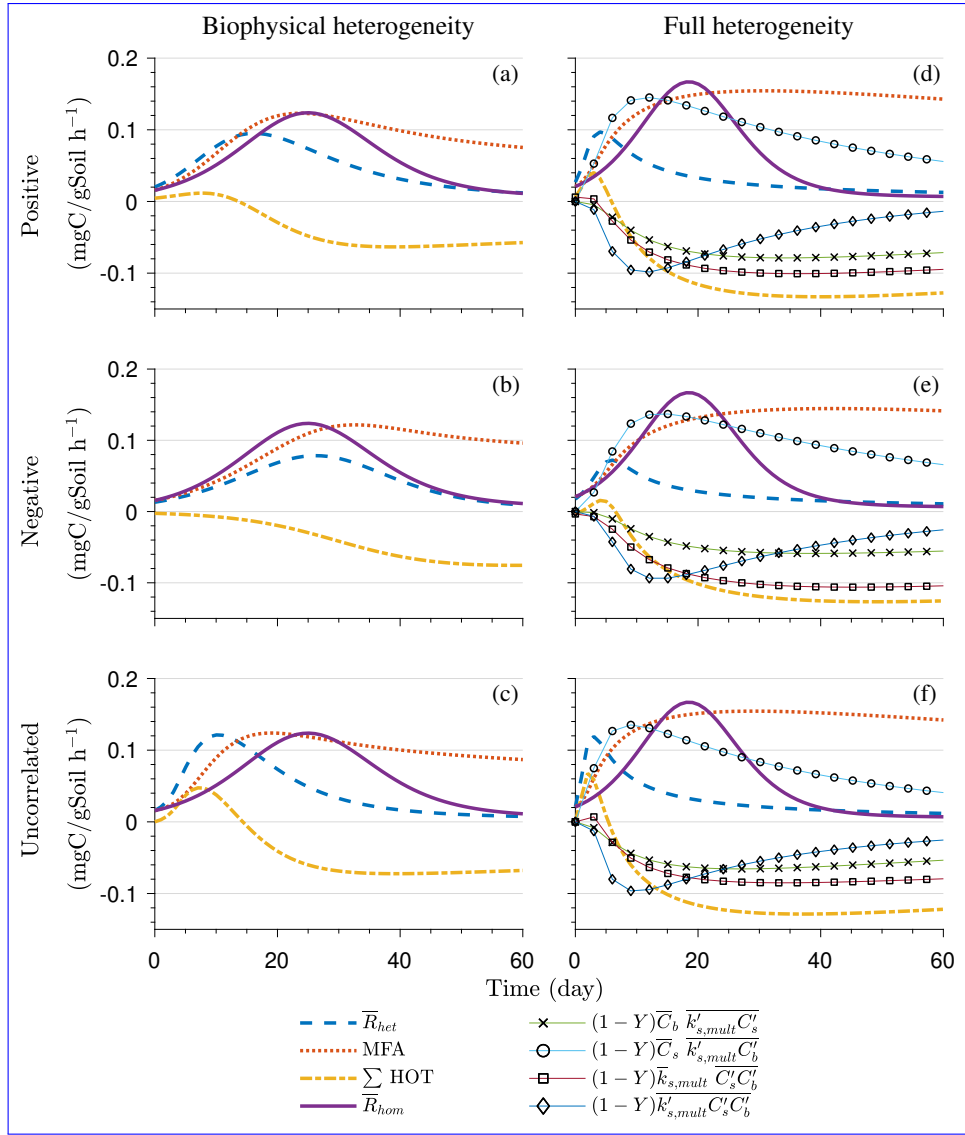


Figure A4. Scenario 2 (transient dynamics with Michaelis-Menten kinetics): temporal evolution of mean respiration rate in the biophysically-heterogeneous system (\bar{R}_{het}), including which includes the mean-field approximation (MFA) and second order terms, and the respiration rate in the homogeneous system (\bar{R}_{hom}), for (a-c) positively the biophysically and (b-d-f) negatively correlated the fully heterogeneous system with positive (a-b), or negative (e-d) uncorrelated and un-correlated (e-f) substrate and microbial C, for multiplicative kinetics.

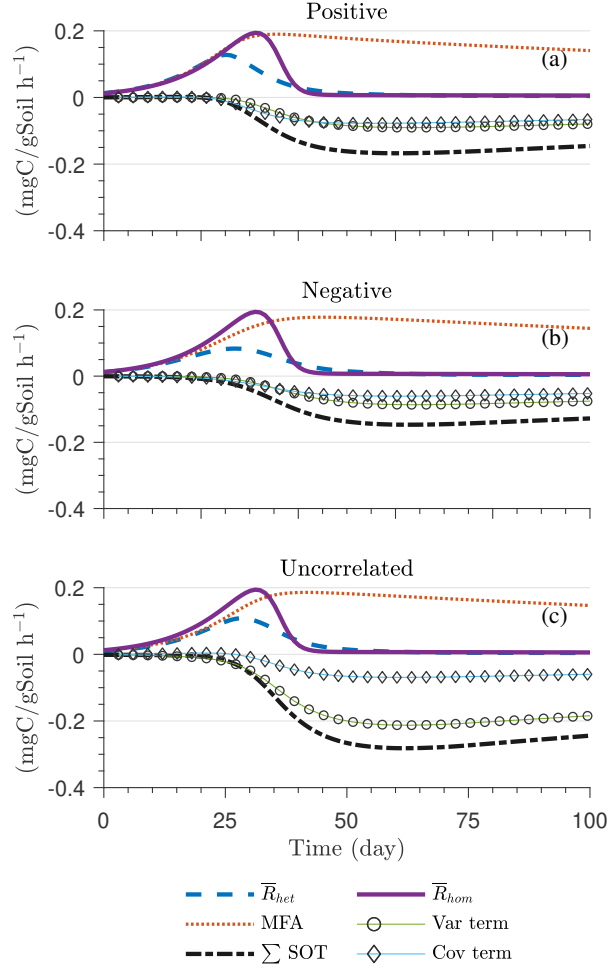


Figure A5. Scenario 2 (HS with Michaelis-Menten kinetics): temporal evolution of mean respiration rate in the biophysically heterogeneous system (\bar{R}_{het} , including the mean-field approximation (MFA), and second order terms), and the respiration rate in the homogeneous system (\bar{R}_{hom}), for (a) positively and (b) negatively correlated, or (c) uncorrelated substrate and microbial C.

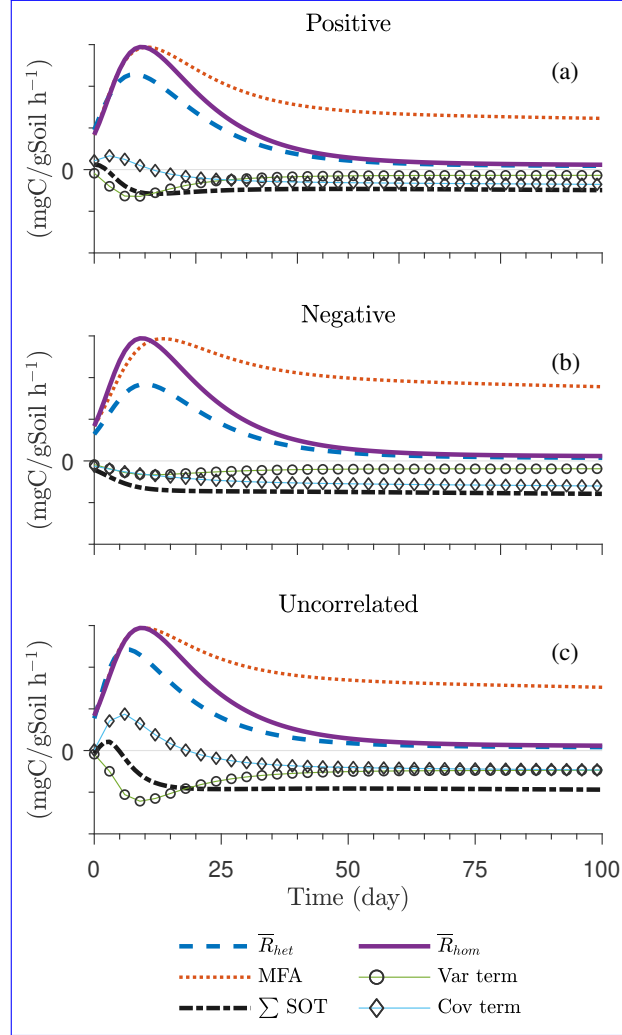


Figure A6. Scenario 2 (transient dynamics HS with inverse Michaelis-Menten kinetics): temporal evolution of mean respiration rate in the sum of biophysically heterogeneous system (\bar{R}_{het} , including the mean-field approximation (MFA), and second order terms for), and the fully heterogeneous respiration rate in the homogeneous system (\bar{R}_{hom}), for different degrees of correlation between the initial (a) positively and (b) negatively correlated, or (c) uncorrelated substrate and microbial C. Only the sum of second-order terms (ΣSOT) is shown because of large number of spatial moments involved.

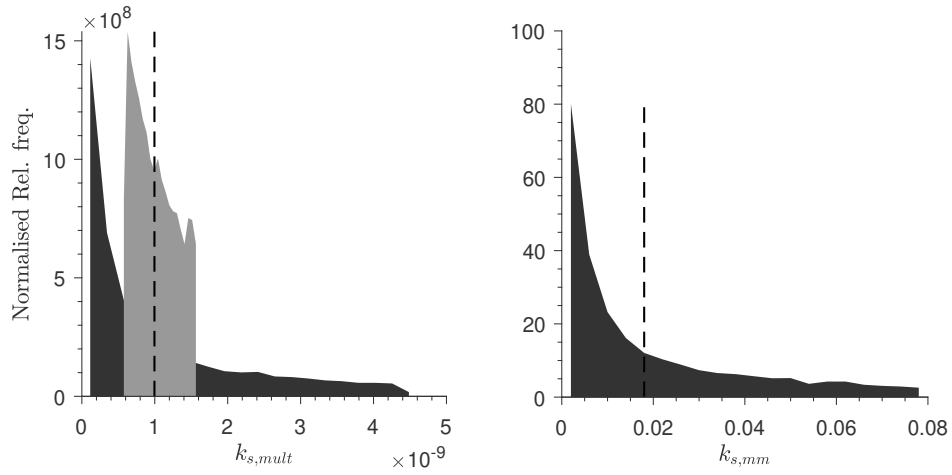


Figure A7. Distribution of the decomposition rate constant for different degrees of biochemical heterogeneity, and for (a) multiplicative and (b) Michaelis-Menten kinetics. Black and grey shadings represent higher and lower degree of biochemical heterogeneity respectively, and the dashed line represents the mean rate constant for the homogeneous system. The half saturation constant K_M \sim K_{MM} is uniformly distributed, not shown in figure.

Table B1. List of parameters. Values in the brackets correspond to the units reported in the brackets.

Parameter	Value	Unit	Description
I	6.06×10^{-4}	$\text{mgCg}^{-1}\text{soil h}^{-1}$	rate of input of external carbon
K_{MM}	5156250 (25)	fgC/grid cell $(\text{mgCg}^{-1}\text{soil})$	half saturation constant (MM)
k_{MM}	0.018	h^{-1}	decomposition rate constant for the MM kinetics
K_{IMM}	2×10^6 (9.69)	fgC/grid cell $(\text{mgCg}^{-1}\text{soil})$	half saturation constant (IMM)
k_{IMM}	0.0045	h^{-1}	decomposition rate constant for the IMM kinetics
$\underline{K}k_M$	7.45×10^{-10} (1.53×10^{-4})	$\text{h}^{-1} (\text{fgC/grid cell})^{-1}$ $(\text{h}^{-1} (\text{mgCg}^{-1}\text{soil})^{-1})$	decomposition rate constant for the Multiplicative kinetics
k_B	0.00028	h^{-1}	decomposition rate constant for the MM kinetics
Y	0.31	h^{-1}	g/cm^3
ρ_{BD}	1.65	g/cm^3	Soil bulk density
ρ_{OM}	1.1	g/cm^3	Organic matter density

Table B2. Initial mean substrate and microbial C in scenarios one and two (in fgC/grid cell); values in brackets are expressed in $\text{mgCg}^{-1}\text{soil}$.

#	Scenario	Initial \overline{C}_s	Initial \overline{C}_b
$\text{fgC/grid cell}(\text{mgCg}^{-1}\text{soil})$			
1	Steady state IC	$1.212 \times 10^5(5.9)$	$2.005 \times 10^5(0.9725)$
2	Transient IC	$2.5 \times 10^7(121.21)$	$2.5 \times 10^5(1.21)$

Table B3. Probability distributions of the parameters for the multiplicative and MM kinetics models. Values in brackets indicate the minimum and maximum parameter values.

		Biochemical heterogeneity 1	Biochemical heterogeneity 2
Multiplicative	k_M (h^{-1}) (fgC/grid cell) $^{-1}$	$\log_{10}\text{Uniform}(-10.1, -8.56)$	$\log_{10}\text{Uniform}(-9.4, -8.9)$
MM	k_{MM} (h^{-1})	$\log_{10}\text{Uniform}(-1.098, -3)$	
	K_{MM} (fgC/grid cell)	$\text{Uniform}(0.25, 49.75)$	
IMM	k_{IMM} (h^{-1})	$k_{MM}/4$	
	K_{IMM} (fgC/grid cell)	$\text{Uniform}(1, 18.4)$	

Author contributions. All authors conceived the initial conceptualization. Arjun Chakrawal and Stefano Manzoni developed the theoretical formalism, performed the analytic calculations and the numerical simulations. Arjun Chakrawal took the lead in writing the manuscript. All authors discussed the results, provided critical feedback, and revised the manuscript.

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