

We thank both reviewers for their time and effort to review our paper. Below we address each of the reviewer concerns separately, with their text in italics and our responses in normal font.

Reviewer #1 (B. Guenet)

We thank the reviewer for the positive comments on our manuscript. Regarding the few minor comments:

1. *The evaluation made by the authors is quite qualitative because of the difficulty to find sites where all the processes represented are measured but I would appreciate a better justification for that (even if I understood why it has been done this way).*

We agree with the reviewer, and therefore have discussed this issue in several places in the manuscript: page 838, lines 17-25; page 840, lines 12-28; page 841 lines 1-4.

2. *I appreciate the discussion but I believe that some lines are missing to discuss about the risk to use this kind of models, with several parameters, in ESMs. Indeed, tuning or optimization possibilities are quite high with an increasing number of parameters and I believe that a particular attention must be paid to ensure that parameters values make sense even after tuning and/or optimization. It is a personal opinion but I would prefer a model that does not perfectly fit the data but with parameters measured or at least in the range of the parameters measured instead of a perfect fit with totally stupid values for parameters.*

We agree with the reviewer's sentiment that care must be taken when choosing parameters for complex models. Such an approach is necessary for ensuring reasonable model predictions, particularly in situations outside of where the model has been tuned. We discussed this point on page 878, lines 17-25.

3. *P824 l5: I did not fully understand what the authors mean with 'unfolding capability'*

This term is only applied to peptides/proteins (Table 2), which under certain conditions can change their structure and therefore have different mineral surface interactions.

4. *P825 l14: 'we did not include the effects of pH', do you mean that $g(pH)$ is fixed to 1 in equation (2)? Please clarify*

Yes, we have clarified this restriction in the text.

5. *P826 equation (4), (5) and (6), why there is some minus after the '=' sign?*

As substrate is consumed (i.e., $dC_i/dt < 0$), CO₂ production increases ($dCO_2/dt > 0$).

6. *P827 l10: BA is expressed in mgC-wet-biomass L⁻¹ but litre of what soil, or soil solution?*

We have clarified the answer to this question in the text (i.e., soil solution).

7. *P833 l7: There is no Fig 3a in the figures section please modify*

This reference should just be to Figure 3; the problem has been fixed.

8. *P834; Section 3.2, there is no words about interaction effects between parameters, are all the effects only additive? The figures suggest that interaction effects may exist. Please present these as a result and add few words in the discussion.*

Yes, interactive effects exist, and that is why we performed concurrent manipulations of both the mineral interaction constants and microbial growth rates (p 834, Line 25-38). The interaction effects are also described in the Discussion section.

9. *P835 l10: This sentence suggests that comparison with 14C data will be done and no data are represented in the corresponding figure. It is a bit confusing, please clarify.*

We have changed this first sentence to clarify this confusion. The text in that paragraph indicates that the simulations are qualitatively consistent with several measurements that are cited in the paper (Trumbore et al. 1995; Baisden and Parfitt 2007).

10. *Fig 4a. The legend is not clear, it represents microbial biomass but why there is lignin etc?*

That is an error in the figure caption; the legend is correct. We have corrected the figure caption.

We thank Reviewer #2 (M. Braakhekke) very much for the thorough review and helpful comments and our manuscript. We address the comments below, with reviewer comments in italics and our responses in normal font.

Reviewer #2: M. Braakhekke

General Comments

1. *One thing I'm missing is discussion (and possibly also model results) related to the priming effect, i.e. the enhanced decomposition of old, autochthonous material when fresh material is added due to simulation of microbial activity. This has repeatedly been put forward as grounds that first-order kinetics models do not fully capture the correct dynamics of SOC (cf papers by Wutzler et al. 2008, Fontaine & Barot 2005). Since the main aim is to derive a more "fidelitous" description of SOC cycling, I think this should at least be discussed. Furthermore it seems to me that the model is capable of simulating such effects so perhaps a simulation experiment would be interesting. This is up to the authors, however.*

Unfortunately, a description of priming that resulted from the perturbations we perform is a substantial amount of work, and beyond the scope of this paper. It is a topic that we are very interested in and relevant to BGC-climate feedbacks, and we envision subsequent papers using this model structure to examine the role of priming.

Methods

2. *The model description is not sufficiently detailed to be fully understandable which makes it difficult for the reviewers to check the validity. I would like to see a list of the mass balance equations including all terms in an appendix or online supplemental material. I think the authors should strive to make the model reproducible by the reader as much as possible. To my mind a journal like GMD is exactly the place for a more exhaustive model description. As mentioned in section 2.1 some things are described in the technical guide of TOUGHREACT. That's fine, but I would at least like to know the boundary conditions applied to the soil carbon model.*

As the reviewer mentioned, there is a detailed TOUGHREACT technical guide; that document, available online, is 251 pages long and gives substantial detail regarding how the mass balance is calculated in the model, as well as referencing some of the many studies this solver has been applied to. The TOUGHREACT code is essentially a reactive transport solver to which various reaction networks and processes can be added. Section 2.7 gives a description of the water and carbon boundary conditions used in the model.

3. *I wonder if the equations related to decomposition presented in sections 2.3 were derived specifically for this model or if they are based on previous work. They are quite complex so I suspect the latter, but I don't see any clear reference. Some of the formulations are difficult to understand, particularly those related to depolymerization. In principle this is not a problem if a reference to a more comprehensive description is included. If the model equations are new a more comprehensive derivation should be presented.*

These relationships have been described in a previous paper from our group ([Maggi *et al.*, 2008]), and we moved that citation to the top of this section to clarify that connection. Regarding the depolymerization relationships, the relationship is simply derived from stoichiometric constraints based on the polymer and monomer stoichiometries, efficiency, and OR ratio. We have added a comment to clarify this issue.

4. *As far as I understand only aqueous transport is considered, i.e. the solid and adsorbed pools are not subject to transport. However, I believe that on the time scales of the simulations in the paper also transport of the solid components is relevant, particularly due to bioturbation. I would expect that bioturbation is an important process in grassland sites from which the measured profiles were collected. Modifying the model and redoing the simulations is not necessary, but I would like to see it mentioned in the discussion.*

Yes, this is a good point, and we have added a discussion of this omission to the first Discussion subsection. We also cite Braakhekke *et al.* (2013), where it was shown that bioturbation was negligible in the forest soil they studied.

5. *Page 822; lines 19–23: the description of z is missing.*

z is depth, and we have added that description here.

6. *Page 824; lines 3–8: can you explain how each property used to group the compounds relates to the processes (decomposition, transport)?*

We have added several sentences to this section to clarify how the properties were used to group the compounds, as suggested by the reviewer.

7. *Figure 1: It is a bit confusing that woody litter, leafy litter, and root exudates are represented by rounded rectangles while they are in fact not pools but input fluxes. I would suggest using labelled block arrows instead.*

Good idea; we have re-drawn the figure in that way.

8. *Figure 2: Can you please make the link with table 2 more clear, for example by adding the abbreviations in an additional column in table 2?*

As suggested, we have changed Figure 2 to have the full compound names, as they are given in Table 2.

9. *Table 1: several comments: (1) I think this table should be placed after table 3; (2) Please explain better in the caption what the columns mean; (3) I think “S1” in the last row should be “S10”*

(1) We have moved Table 1 to be after Table 3 and re-numbered them in the text; (2) we have added text to the Table 1 caption to explain the multiplier factors and the last two columns; (3) that (S1) was an error during typesetting, and has been corrected.

10. *Page 824; line 23: “encapsulated in aggregates”. Elsewhere it is mentioned that the effect of aggregates is not considered in the model.*

We have corrected this error.

11. *To my mind sections 2.4 and 2.5 should be placed before section 2.3. Sections 2.4 and 2.5 introduce the decomposition reactions for monomers and polymers, and 2.3 deals with the rates of these reactions, and how they affect the different species. In fact, some of the symbols used in section 2.3 (Y_i , x_i) are not explained until the later sections.*

We moved those sections, as suggested.

12. *I cannot find the mathematical formulation for the production of the carbon pools. Fig. 1 shows that a part of the decomposition flux of donor pools flow to other pools, and Fig. 2 shows these partitioning fractions. However I can't trace this to the mathematical equations. I would expect the quantities displayed in Fig. 2 to show up in the mass balance equations somewhere.*

The end of section 2.2 describes the carbon transformation pathways and how the rates are calculated. The carbon pools are consumed using equation 2 (i.e., the kinetic reaction rate expression) and produced from microbial death (equation 6; Figure 2) and carbon inputs (Figure 1). The third right hand side term of equation (1) represents these sources.

13. *I believe that the notation in equations (2)-(6) is not completely correct.*

This notation is confusing, as you suggest, although you have interpreted it correctly. We have taken your advice and re-labeled the terms in equations 2-6.

14. *Eq. (7): RO/C varies per compound, right? Please add an i to indicate this.*

Done, as suggested.

15. *Eq. (9): Please replace R_0 with RO*

Done, as suggested.

16. *Page 828; line 3: can you please explain more clearly what x_j denotes?*

When equation (8) is expanded for each compound, the resulting stoichiometric coefficients affect the biomass yield. x_j are those stoichiometric coefficients.

17. *16. Section 2.6: The representation of ad- and desorption is described in insufficient detail. It is only mentioned that “forward (adsorption) and reverse (desorption) rates are imposed”. However, this doesn’t show up in any of the mass balance equations. Furthermore, it is not mentioned how adsorption affects decomposition. I assume that species adsorbed to minerals are protected from decomposition, but this should be clearly stated.*

We have added text to the Methods section to clarify that the sorption reactions are subsumed in the source term of equation (1). We have also added the statement that sorbed species are protected from decomposition.

18. *Page 829; line 26: Can you please explain by “characteristic lengths”? Also, it would be nice to see the root input profiles in a graph, e.g. in Fig. 3 or 4, or in supplementary material.*

By that we meant “exponentially decaying depth profiles with length scales of 1, 7, and 12 cm”, and have revised the text accordingly.

19. *Page: 830; line 2: A minor comment: to my mind checking convergence to steady state based on the first derivative over time is risky. If pools accumulate very slowly it may seem as if they are close to steady state, while in fact they are not. Better is to involve also the second derivative. But I trust that the authors made*

sure there was true convergence.

Yes, we did.

20. *Section 2.8: I agree with the comment of Bernhard Ahrens regarding the delta-notation for ^{14}C . Furthermore, in section 3.3 and 4.1 the model results for ^{14}C are discussed in the context of observations, but the latter are not shown anywhere. It would be helpful to show several ^{14}C profiles (possibly in supplemental material) for readers who are not very familiar with such data, also in view of the unrealistically low delta- ^{14}C values predicted for the topsoil.*

Yes, the editor or typesetters changed the notation from our submitted version, which used the notation $\Delta^{14}\text{C}$. We will ensure that is corrected in the published version. Because we did not have sufficient data from any site with measured $\Delta^{14}\text{C}$ values to reasonably test our model, we do not want to reproduce previously described radiocarbon values. However, we did cite publications with $\Delta^{14}\text{C}$ values so readers can find examples if they are interested.

21. *Section 2.9: The two experiments described in lines 10–20 are not fully clear to me. For the first experiment it is stated that “we doubled all chemical species initial concentrations from those at the end of the 10000 yr simulation, and performed a 500 yr simulation”. For the second experiment it is stated that “we performed pulse carbon input experiments by doubling the steady-state concentration of all compounds in seven depth intervals (0–10, 10–20, 20–30, 30–40, 40–75, 75–125, and 125–200 cm)”. I don’t really see the difference between the two experiments.*

The first experiment doubled concentrations between 0 and 20 cm, while the second doubled values in the distinct layers mentioned. We used the first simulation to examine transient responses in detail, and the second series of simulations to characterize effective turnover times as a function of depth, and how those depth intervals interact with carbon transformations in other depth intervals.

Results

22. *Could you perhaps also give some numbers to the average predicted DOC fluxes (or show a graph in supplementary material) and compare with observations from previous studies? A good reference for the latter could be Kindler et al., 2011 (GCB). It is mentioned that since DOC concentrations are very variable in time, they is not a good metric for comparing with predictions. However, I believe that time-averaged fluxes (such as those presented by Kindler et al) could be a good reality check for the model, at least in terms of order of magnitude.*

Thank you for this idea. We extracted the DOC flux from our model, and on an annual basis it is $6.2 \text{ g m}^{-2} \text{ y}^{-1}$, which matches the Kindler et al (2011) value for grasslands of $5.3 \pm 2 \text{ g m}^{-2} \text{ y}^{-1}$ very well. We have added this comparison to the paper in the Results section.

23. *Page 833, line 13: the part starting from “where most of...” is a bit confusing. I would suggest writing “which receives most of the input...” or similar.*

Done, as suggested.

24. *Figure 6: Please consider making this figure bigger and omitting the errorbars for the observations.*

We want to show all the sensitivity scenarios in one figure so that the impacts can be compared, and the error bars are valuable to distinguish, at least qualitatively, how significant each specific parameter perturbation was.

25. *Page 834, line 3: “0 percent microbial biomass below 40cm depth”. I assume the biomass it’s not actually zero since this would mean there is no decomposition and carbon stocks would grow very large. Or is everything removed by transport?*

The sentence is referring to the proportion of biomass that is fungal, not total biomass content.

26. *Page 834, line 28: Please replace “-50cm” with “0-50cm”. Same for page 836, line 27.*

These were typesetting errors; we will ensure they are both corrected.

27. *Page 835, line 18: The sentence starting with “Using...” is difficult to follow. Consider revising.*

We added a phrase to indicate that we were referring to a first order loss term.

28. *Figure 7: Like Fig. 6, please consider making these figures bigger, possibly by moving some to supplementary material*

We feel it is important to keep these figures together, since they in combination show

the dynamics of the system.

Discussion

29. Section 4.1.1: near the end of the section it seems as if the aim is to fit the observations as close as possible (“mostly outside standard deviations”, “biases”). However, given that the observations come from many different sites, while the model is only run for one and is not calibrated, I guess they are only included for comparison in terms of order of magnitude.

Yes, that is correct.

30. Page 844, line 1: please add an “s” after “move”.

The sentence is correct as is.

31. Page 846, line 27: I believe the correct spelling is “in silico”.

We removed that phrase.

We thank B. Ahrens for his insightful comments on our manuscript, and attempt to address his three points below (his comments in italics and our responses in normal font).

- 1. You very openly discuss that under the current parameterization your model gives negative $\Delta^{14}\text{C}$ values in the first centimeters (P.835–L.12; P.839–L.27). Visual inspection of Fig. 6 d-f would suggest $\Delta^{14}\text{C}$ values of $-100\pm 25\%$ in the first centimeters. I would have expected that the modeled $\Delta^{14}\text{C}$ in the first centimeters would easily reflect that litter inputs have had a $\Delta^{14}\text{C} > +69\%$ from 1957–2003 (“bomb-peak”). Could you elaborate which mechanisms in the model are right now responsible for negative $\Delta^{14}\text{C}$ values in the first centimeters, corresponding to conventional ^{14}C ages of around 900 years BP? Sorption processes? Very fast turnover of litter inputs? Could that also be related to the vertical resolution of the model?*

As you mention, the model predictions did not match expected enriched $\Delta^{14}\text{C}$ values in the near-surface soil, which we believe indicates underlying mechanisms not represented in the model. As we discuss in the Discussion section:

“However, for context, we estimated that an increase of about 30 percent in SOC concentrations resulting from plant inputs over the past several decades would lead to a close agreement with observations. Thus, an additional young and protected carbon pool of small size (Fig. 3), and effectively not in equilibrium with the aqueous phase, can explain the difference between our predicted and commonly observed $\Delta^{14}\text{C}$ values near the soil surface. Our model allows for an additional non-equilibrium carbon pool that could be tuned to match these $\Delta^{14}\text{C}$ and SOC profiles, but we have avoided that type of tuning here. Processes that may be good candidates for this level of protection include aggregation and formation of colloids, which have been shown to substantially affect chemical mobility and carbon decomposition rates in soils (Daynes et al., 2013; Kausch and Pallud, 2013; Six et al., 2000).”

To clarify these points, we have indicated in the Results section that this discussion follows.

- 2. Throughout the text you use the $\Delta^{14}\text{C}$ notation, but the $\Delta^{14}\text{C}$ notation in Fig. 6d-f. Is this by accident? In my opinion, the $\Delta^{14}\text{C}$ notation should be preferred because it is independent of isotopic fractionation (Stuiver and Polach, 1977). Because one probably does not include isotopic fractionation due to photosynthetic fixation and microbial processing into the model, the $\Delta^{14}\text{C}$ notation should be more appropriate for model output.*

It was not by accident, but by error from the typesetters. Our submitted text used the $\Delta^{14}\text{C}$ notation, and we will ensure that this problem is corrected in the final version of the paper.

- 3. The distribution coefficient K_d is very helpful to get an idea about the sorption affinity of the different compounds (Table 2), you note, however, that you use a*

dynamic approach because of competing sinks and sources (e.g., microbial consumption). How do the adsorption and desorption rates k_f and k_r compare to the maximum specific consumption rates μ_i ?

In the baseline version of the model, k_f is $6.6e-8$ per second and k_r is scaled by the factors shown in the last column of Table 2. The maximum specific consumption rates are given in Table 3. Note that this comparison is not particularly helpful to understand the relative rates between these processes, since the maximum specific consumption rates are modified by the Michaelis-Menten kinetics (equation 1) and are therefore often much lower than the maximum values. We have added text to Section 2.6 to clarify the sorption rate values.