

Interactive comment on “Biogeochemical model of CO₂ and CH₄ production in anoxic Arctic soil microcosms” by Guoping Tang et al.

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Comment 1: Community land model carbon nitrogen (CLM-CN) predominantly represents aerobic decomposition of SOM. In this manuscript, authors propose to include anaerobic processes in this model by integrating new experimental data for redox potential, pH, and temperature parameters from Arctic soils. This manuscript is very thorough. It's amazing to see parameterization of model with experimental data! While this work has some flaws, it is a huge step forward in closing the gap between modeling and experimental data integration. I'm impressed by the author's knowledge of biogeochemical processes in soil and effort to connect real world mechanisms to the modeling results; this is no small feat. It is clear they gave a great deal of thought to their results.

C1

Response 1: Many thanks for the compliments and very nice constructive comments.

Comment 2: In general, I would recommend the author's provide stronger justification for determining that the most limiting factor for SOM turnover is hydrolysis of macromolecules. This both served as the foundation of this work and is continually provided as an explanation to observations. While it's tough to cover all possible scenarios in soils, authors should address other potential factors that could drive the rate of SOM turnover and justify why they believe hydrolysis of macromolecules is the most limiting factor.

Response 2: We agree that hydrolysis is a limiting factor. We also agree with referee #1 that it is controversial to state that hydrolysis is the most limiting factor. At least, the evidence from the data referred in this work does not unequivocally substantiate the statement. As a result, we try to be balanced and discuss about possible new data needs to better understand and quantify hydrolysis.

All the referees comment on the need to mention other factors. In response, we make revisions to clarify the scope of this work, to put our work in the context of comprehensive hydrologic, geochemical and biologic processes that control soil carbon mineralization, and describe using 3-D high resolution grids to account for heterogeneity, and CLM-PFLOTTRAN to use reactive transport models to improve the mechanistic representation in land surface models. Please see response to other referees for more details.

Comment 3. In the conclusion, I think it would be nice for the author's to add some suggestions for parameters/processes that could be incorporated into this model in the future or specific geochemical measurements that experimentalists should consider collecting during their studies.

Response 3: These are nice suggestions. As mentioned in Response 2, we add discussions about next steps.

C2

—Specific comments—

Comment 4: P3L10-11: "... the hydrolysis and fermentation reactions have been poorly quantified." I'm not sure I follow the point being made here. Is this suggesting that hydrolysis/fermentation of SOM is poorly quantified (in general) or specifically in arctic soils?

Response 4: add "represented and quantified in Arctic as well as temperate and tropical soils" to clarify the point.

Comment 5: P4L28-29: What is a "low-center polygon"? It is frequently referred to in the text of this article, yet it is unclear to me what it is. This seems like site-specific terminology that may be worth describing. I'm not sure how many readers would know what this is. I'm also assuming the "center" sampling location is a slope since the other two are the "ridge" and "trough"?

Response 5: add "(a typical arctic geographic feature in the low lands with soils surrounded by ice wedges, see cited references for more information)".

Comment 6: P7L28: What do SOM3 and SOM4 represent? LabileDOC, SOM1, SOM2 and the biomass pools were described, but not SOM3 and SOM4. Furthermore, SOM4 isn't included in the fractions listed on P7L29. Is it supposed to be included in this list of fractions? If not, then why is it excluded?

Response 6: SOM3 and SOM4 are like SOM1 and SOM2, two additional soil organic matter pools in CLM-CN (Fig. 1). We add "(the rest is assumed to be SOM4, e.g., $f_{SOM4} = 1 - f_{LabileDOC} - f_{SOM1} \dots$)"

Comment 7: P8L1-2: The turnover time of SOM3 and SOM4 are not listed – these fractions need to be better described or explain why they are excluded.

Response 7: add "(as the turnover time for SOM3 and SOM4 are 2 and 27 y, respectively, Fig. 1)".

C3

Comment 8: P8L7-9: Nice explanation for "back of the envelop" biomass estimation

Response 8: Thanks.

Comment 9: P10L26-27: Are there other potential reasons why the rate of CO₂ would stabilize? Limitation of some other resource? For instance, N? Does this study have evidence to support that rate of CO₂ respiration stabilized because of hydrolysis of polymers?

Response 9: These are very good questions. As we mention ahead of section 2.1, "While nitrogen (ammonium and nitrate) concentrations can affect carbon mineralization (Lavoie et al., 2011), we do not account for this effect because of a lack of nitrogen measurements from these experiments." As we mention earlier, we do not have specific direct evidence to support polymer hydrolysis as the limiting factor.

Comment 10: P11L8: parameter $Fe_3 = 0.02$ is above the max value in the range of observed values stated on P8L14, can the authors comment on why they might need to increase this value beyond observed values to help the model better match observations for Fe(II)? Do you have any suggestions for some other parameter that should be included or other parameter values that could be altered to help achieve a better model fit, while maintaining values within experimentally observed value range?

Response 10: The observed range is from another site. It is not directly applicable here. In the revision, we revise from "we start with $f_{Fe3} = 0.005$ " to "While bioavailable Fe(III) in soils is not well defined (e.g., Hyacinthe et al. 2006; Poulton and Canfield 2005), we start with $f_{Fe3} = 0.005$ and evaluate the impact with a range of values."

Comment 11: P11L11-14: How do these model observations relate to experimental data? Is there any experimental evidence (either from your original work or other soil Fe literature) to support that as Fe₃ increases there is a decrease in CH₄ resulting from competition between methanogens and iron reducers? Why wouldn't this also be the case when $Fe_3 = 0.01$?

C4

Response 11: We add “(rather than strict thermodynamic control, e.g., Bethke et al., 2011; direct inhibition, e.g., van Bodegom et al., 2004; or indirect inhibition through substrate competition, e.g., Mill et al., 2015, Reiche et al. 2008)”. As discussed in these cited references, Fe reduction is known for inhibition of methanogenesis.

Comment 12: P11L29-31: This statement contradicts L25-26. L25-26 states as pH increases, CO₂(aq) increases. L29-31 states as pH increases, CO₂ (aq) decreases. Please provide an explanation.

Response 12: This was because CO₂ in the aqueous phase here means a specific aqueous species rather than total CO₂. To avoid this confusion, we add (aq) after CO₂ and the sentence is revised from

“As the pH increases above the carbonic acid pK_a (around 6.3 at standard condition), CO₂(g) in the headspace and CO₂ in the aqueous phase decrease as HCO₃⁻ becomes dominant, and the gas-phase fraction decreases dramatically.” to

“As the pH increases above the carbonic acid pK_a (around 6.3 at standard condition), CO₂(g) in the headspace and CO₂(aq) species decrease as HCO₃⁻ becomes the dominant species in the aqueous phase, and the gas-phase fraction decreases dramatically.”

Comment 13: P12L19: I keep having to look back at what “WEOC” means. I would recommend using some other terminology. Also, this sentence should reference Table 2 not Table 1.

Response 13: As suggested by the other two referees, we spell out WEOC. The table reference is corrected.

Comment 14: P12L20-22: Is this comparable? The values for rapid CO₂ release in Figure 4 look nearly double or triple the observed values. It appears that CO₂ values for organic center at a LabileDOC = 0.02 fit the experimental data best out of all of these scenarios.

C5

Response 14: revise to “... the underprediction of the early CO₂ increase in the headspace are more or less mitigated.”

Comment 15: P12L29: “high center polygon trough”? I thought “center” and “trough” were two different sampling sites? Please clarify and be consistent throughout the paper. Same error P13L6.

Response 15: revise from “. . .from the high center polygon trough” to “. . .from a trough location in a high center polygon. . .”

Comment 16: P13L19-20: I don't follow – how do these studies demonstrate that hydrolysis of macromolecular organics by extracellular enzymes is the rate limiting step? What about bioavailability? Limitation of some other resource?

Response 16: As we discuss earlier, we do not have direct unequivocal evidence for this.

Comment 17: P13L24-26: Please rewrite this sentence for clarity.

Response 17: remove “(or produce substrate for)” and add “in the slabile = 0.2 case”. The sentence reads:

“With slabile = 0.2, the model generally predicts less CH₄ and more CO₂ than the case with slabile = 0.4 because less SOM is assumed to respire through the anaerobic pathway in the slabile = 0.2 case (Fig. S5).”

Comment 18: P13L31: “the model substantially underpredicts: : .:” Please include a figure number.

Response 18: include figure number: Fig. 4b3.

Comment 19: P14L1: It could also be attributed to populations at that particular site grow more rapidly than the populations at other sites. Hard to say without a T₀ measurement: : : I would tread lightly with this, you don't have strong experimental evidence to support this statement.

C6

Response 19: remove “, indicating possible high initial abundance”

Comment 20: P14- first paragraph: The text says the opposite of what is demonstrated in Figure 5.

Response 20: The legend was wrong. It is corrected. The numbering for the subplots was moved to the right corner to avoid overlap with the legend.

Comment 21: Figure 5 shows the lower initial biomass results in more CH₄, FeII, pH increase, etc. Is it possible the figure legend is wrong?

Response 21: See Response 20

Comment 22: P14L10-12: OK, but if the OM soils are better buffered why are there rapid changes in pH for both the observed and experimental data for OM soils? Figure S6. OM soils appear to have rapid pH changes occur sooner than mineral soils, despite buffering? Please explain.

Response 22: The initial drastic drop in pH for OM soils are due to the fermentation of a large amount of initial labile carbon. Because of the abundance of simple substrates, Fe reduction and methanogenesis rates are high later, resulting in fast pH increase. It is really a complex nonlinear relationship.

Comment 23: P16L21: change “enhancing” to “enhances”

Response 23: revised.

Comment 24: P16: Transparent science! Thanks for making your code and data available!

Response 24: You are welcome. We are happy to share.

Comment 25: P17: It's unclear what a pH response and temperature response function are. Please better define. What is the reader supposed to take away from this information?

C7

Response 25: add “(reaction rate adjustment factor as a function of pH)” and “(reaction rate adjustment factor as a function of temperature)”. As we discuss in the introduction and results and discussion sections, the take-away is that these two response functions are an important source of uncertainty.

Comment 26: All tables and figures should be able to stand on their own. Improve caption text and add full legends (colors, symbols, and patterns defined in each figure).

Response 26: improved. Please see the marked manuscript in response to referee #1.

Comment 27: -Please format Table 2.

Response 27: this is reformed (see page 27).

Comment 28: -Figure 2 caption L5 add “as” after “such”

Response 28: added.

Comment 29: -Figure 5 caption text does not match figure. Legend suggests lowest initial biomass results in highest CH₄. Please make full legend visible (partially covered up).

Response 29: See Response 20.

Final marked revised manuscript is available in response to referee #1.

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C8