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Discussions

Interactive comment on "Biogeochemical model of CO_2 and CH_4 production in anoxic Arctic soil microcosms" *by* Guoping Tang et al.

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Comment 1: Soil carbon models are a critical source of uncertainty in Earth system models both due to limitation in model-data and process representation. This manuscript addresses modeling hurdles in a key process (anaerobic decomposition) missing in most soil car- bon models. This work is a timely, novel, and carefully conducted. However I am uncomfortable with the current introduction, justification, and implication presented in the manuscript. The actual results section is very strong but would suggest extensive revisions to the introduction and conclusion.

Response 1: We appreciate that the referee spent valuable time in reviewing this manuscript, and provided very constructive comments from a different perspective. As detailed below and highlighted in the revised version, we made extensive revisions

to improve the introduction, justification, and implication components as the referee suggested, with a focus on clarifying the scope of this work, and putting it in the comprehensive context of earth system model concisely. We hope the manuscript is substantially improved.

Comment 2: In general, I would suggest better connections between and within the introductory paragraphs. The main paragraphs jump around and paragraphs themselves lack coherent structure.

Response 2: This is because that we tried to avoid lengthy discussion about the context of carbon mineralization and methane production, consumption, and transport. Besides extensive revisions in the introduction as highlighted in the attached marked revised manuscript, we add the following paragraph in the end of the introduction section to concisely describe the context and limit our scope of this study, which reads:

"The carbon cycle involves coupled hydrological, geochemical, and biological processes interacting from molecular to global scales. The implicit empirical first order approach used in existing LSMs limits our understanding of the land atmosphere interaction and is a source of prediction uncertainty. To improve our understanding and reduce prediction uncertainty, we attempt to use relatively more explicit mechanistic representations developed in the reactive transport model literature (Tang et al., 2016). Even though explicit representation does not necessarily improve the match between the predictions and observations over well-tuned existing models immediately (e.g., Wieder et al. 2015; Steven et al. 2006), our approach provides a systematic means to incorporate on-going process-rich investigations to improve mechanistic representations in LSMs across scales. As a preliminary study, we constrain our scope to extending CLM-CN with minimum revision to describe anaerobic CO2 and CH4 production from several recent microcosm studies in this work. We discuss next steps briefly results and discussion section." We also add the paragraph in the end to describe future directions:

"As the experimental data from Roy Chowdhury et al. (2015) and Herndon et al. (2015) are shown to be invaluable, iteration of further biogeochemical modeling and new experiments that span a range of temperature, pH, redox conditions, and include detailed characterization of hydrolysis, microbial and enzymatic activities will further improve mechanistic understanding and representation of carbon mineralization and methanogenesis. The oxidation of methane, Fe(II), etc., may be equally important for simulating the carbon cycle. The biogeochemical model is typically coupled with a hydrological model to account for heterogeneity in structured soils using 3-dimensional high resolution grids. As CLM-PFLOTRAN couples CLM with a reactive transport code PFLOTRAN, these hydrobiogeochemical model developments can be directly incorporated and tested from laboratory to global scales. As we implement processes, such as gas and aqueous transport through soils and aerenchyma, explicitly representing microbial processes for carbon decomposition, incrementally, we hope the new modeling framework will be more and more useful for future investigation and land surface model developments."

Comment 3: I would also like to see a discussion/acknowledgement of other mechanisms that could influence anaerobic decomposition which were not captured by the proposed model. While not ALL processes can be included in a model, some acknowledgement of those missing processes and either how then can be incorporated into future work or how they influence current simulations are appropriate and highlight the limitations of the proposed model.

Response 3: the two paragraphs added in response 2 put the current work in the context of other processes. In responses to referee #2,we discuss describing impact of flushing on biogeochemical processes.

Comment 4: Relatedly the authors need to address how these detailed pool based kinetic models scale to well-structured heterogeneous soils. Great detail is gone into on the chemical processes governing methanogensis in the introduction but there is no discussion of how the physical structure of the soil plays into substrate and oxygen

availability and the inherent limitations to applying mechanistic pool models to highly structured soil columns. While this is a common shortcoming of soil carbon models I feel that, given the level of processes detailed covered in the model, this is critical to address and justify utilization of a pool model with such explicit process representation. Minimum the authors need to acknowledge that the scaling of known kinetics from well-mixed experiments to highly structure soil cores is a relevant open question in the field.

Response 4: As in response 2, we add discussion about using high resolution to deal with heterogeneity; in Tang et al. 2016, we actually discussed potential ways to account for oxygen limitation and even oxygen transportation and consumption; While scaling is a grand challenge, there are evidences that the parameters determined in the lab are applicable for field studies in our previous studies (e.g., Tang et al. 2013b). As a preliminary study, we feel it is better to focus on the current scope.

Comment 5: The great strength of this manuscript is the highly detailed and careful analysis of the proposed model, grounded against a data set. This was very well done and I feel should provide the backbone of a new discussion section which could be extended to suggest potential follow up experiments based on the model results. However there is no formal model-data integration nor a comparison with an adequate number of data sets to justify this as a mature component of a new CLM module, as is implied by the current introduction and discussion. If this is the manuscript that the authors want to write then I would suggest more data sets, a formal data-model integration, and demonstration of improvement to previous ratio-based models. But the current analysis would be completely appropriate in a different context which focused more on implications to future experimental designs and long term model projections. I strongly encourage the authors to carefully consider an alternative framing of this very interesting study.

Response 5: We appreciate the referee's positive comments, and very constructive advices. As discussed in response 2, this is a preliminary study with focus on mechanistic representation. We are interested in incorporating more existing data, but are limited by lack of detailed data such as pH, Fe, organic acids, etc., in the existing studies. In the revision, we mention potential next steps and further model data iteration exercises.

–Line by line response–

Comment 6: P2L2 Actually many of the IPCC models suggest that SOC will increase in the northern latitudes due to increases in inputs (Todd-Brown et al 2014), I would suggest softening this statement to reflect the huge uncertainty in the current state of the science. Less controversial would be a statement referring to a general ramping up of the entire car- bon cycle in response to climate change, increases are expected in both primary pro- duction and decomposition. Whether the net effect will be to convert SOC to CO2/CH4 is highly debated.

Response 6: revise ". . . are widely expected to accelerate . . ." to ". . . may . . ." to soften the statement.

Comment 7: P2L2-14 I like the content of this paragraph but it needs to be re-organized. There are three distinct topics in this paragraph which would be better served breaking them up/integrating with later parts of the manuscript: a review of expected high latitude SOC vulnerability to climate change, summary of CLM-CN representation of anaerobic conditions (coupling this with a general review of aerobic decomposition would not go amiss here but that is a soft suggestion), and the comparison to lab incubations.

Response 7: we separate the paragraphs into two, and put our study in the context concisely (see marked revised manuscript).

Comment 8: P2L15 CH4 is critical not just because of it's high global warming potential but also because of it's emissions rate and residence time in the atmosphere. Please add some citations to reflect this.

Response 8: add residence time and production rate (italic) as

"Because CH4 has a 100-year global warming potential that is about 26 times greater than CO2 (Forster et al., 2007;IPCC, 2013), and an atmospheric residence time of approximately 10 years (IPCC, 2013), and methanogenesis rate can be high under favorable conditions, . . ."

Comment 9: P2L19 Why is this lag critical?

Response 9: We rewrite the paragraph as follow: "Methanogenesis is widely parameterized as a fraction of carbon mineralization (Wania et al., 2013; Oleson et al., 2013; Koven et al., 2015; Cheng et al., 2013). However, the ratio of CH4 to CO2 ranges from 0.00001 to 0.5 (Wania et al., 2010; Drake et al., 2009; Bridgham et al., 2013), highlighting limitation of this simplistic empirical approach. The wide range of CH4 to CO2 ratio, also shown as the observed time lag of CH4 accumulation behind CO2 in anaerobic microcosm ranging from days to years (Knoblauch et al., 2013; Roy Chowdhury et al., 2015; Cui et al., 2015; Hoj et al., 2007; Fey et al., 2004; Jerman et al., 2009; Tang et al., 2013c), is due to different temperatures (Fey and Conrad, 2003; Hoj et al., 2007; Jerman et al., 2009; Cui et al., 2015), initial abundance of methanogens (Conrad, 1996; Knoblauch et al., 2013), and the wide range of redox buffering provided by the alternative electron acceptors (Estop-Aragonés and Blodau, 2012; Fey et al., 2004; Jerman et al., 2009; Yao et al., 1999; Conrad, 1996; Knorr and Blodau, 2009). Accordingly, methanogenesis is explicitly represented in some models (Xu et al., 2015; Grant, 1998) and the reduction of alternative electron acceptors is explicitly represented in others (Fumoto et al., 2008; Segers and Kengen, 1998; Van Bodegom et al., 2001; van Bodegom et al., 2000). However, these models do not have an aqueous phase that is essential for explicit biogeochemical calculations, e.g., pH, Eh, and thermodynamic calculations. As the free energy of methanogenesis reactions is less favorable than the reduction of O2, NO3-, Mn (IV), Fe(III), and SO42- along the redox ladder (Conrad, 1996; Bethke et al., 2011), it is important to explicitly simulate the redox condition to accurately predict methanogenesis."

to connect CH4:CO2 ratio with lag. The wide range of CH4:CO2 corresponds with the wide range of lag time.

Comment 10: P2L25 Why is the CH_4:CO_2 ratio important? Maybe lead with this being a critical parameter for current models and then show how this is a dynamic response to the competing Redox ladder. I think this is where the authors are trying to go with this but it is lost in the paragraph. Could the proposed model be compared with the standard ratio model?

Response 10: As in response 9, the manuscript shows that a constant CH_4:CO_2 ratio approach introduces prediction uncertainty, and our mechanistic model has the potential to reduce the prediction uncertainty. We feel it to be premature to conclude whether the mechanistic model perform better than a standard ratio model. The mechanistic model is expected to work better than the empirical for these specific data sets. It is challenging to extend the comparison to other available datasets because the lack of measurements (e.g., pH, Fe, etc.) to support the mechanistic model. Our long term goal is to evaluate if a mechanistic model performs better than a standard ratio model. This is only the first step toward the long term goal.

Comment 11: P2L33 Why is an aqueous phase essential for these calculations? Soil decomposition models are implicit descriptions of carbon dynamics anyway, why do we need an explicit representation of this process? Can a micro scale process like an explicit terminal electron acceptor model be simulated on the macro scale? I would suggest placing this study in the context of the increasing number of ?explicit? soil carbon dynamics models (ex: Wieder et al 2015). These models may or may not increase the overall accuracy of predictive dynamics over a well-tuned traditional model but they can provide critical scientific insights into the process of soil decomposition. This introduction lacks this critical nuance and oversells the capabilities of the proposed model.

Response 11: The aqueous phase is essential in that pH, Eh, thermodynamics, etc., are defined in the aqueous phase. As in detailed in Response 2, we put this study in

the general context in the revision and acknowledge that explicit representation may not necessarily improve the match with the observation immediately over existing well-tuned models.

Comment 12: Sect2.2.1(and elsewhere) Please refer to model pools and other variables by name (variable) consistently throughout the manuscript, ex: organic acid pool (Ac). This reduces the need to refer back to tables/sections. Manuscripts are rarely read linearly and having to search for abbreviation definitions slows down reading.

Response 12: spell out.

Comment 13: P8L20 I applaud the authors for making their scripts available in the supplemental. Thank you.

Response 13: Thanks.

Comment 14: P12 Nice job walking through potential drivers of model-data mismatch. These provide a rich pool of candidates for future investigations. I feel that this should be the main focus of the conclusions. Given that a single data set was used and no formal model-data integration done this model is not quite ready for a full land carbon model integration as is implied by the authors in the introduction and conclusions. What IS done quite elegantly is an analysis of several representations of potential mechanisms and how they influence overall carbon mineralization in the context of a common model structure.

Response 14: Thanks for this nice evaluation. We revise the discussion and conclusion section to further strengthen these points.

Comment 15: P12 WEOC, TOTC are an unusual acronym in the field. Consider writing out the full name instead, I found myself forgetting what it stood for around here and having to go look it up. See previous comment about variable/pool references.

Response 15: spell out.

Comment 16: P13L19 This is a highly controversial statement that does not belong in the results section. While it is appropriate to highlight the relatively low amount of mineralized carbon there are several possible mechanisms for this that are unrelated to the chemical structure as suggested by this statement. Just because that is the explanation that fits into the model that is presented, it is not the only explanation (I'm thinking of various physical mechanisms like co-location and aggregate formation, as well as the substrate rarity argument ala Allson 2006). Please move to the conclusion and soften this statement considerably.

Response 16: revise "... is the rate limiting step ..." to "... could be a rate limiting step..." to soften the statement.

Comment 17: P14L17 Cite the equation reference for f(pH)

Response 17: add (Eq. 3).

Comment 18: P14L26 Given the noise generally inherent in these measurements I would hesitate to call this a substantial difference. Could you can provide error bars for the data or some kind of significance testing.

Response 18: The sentence reads "These differences translate to substantial differences in model predictions". It is model predictions not observations. We add "(Fig. S7)" to avoid confusion. Instead of showing the average with error bars, the duplicate/triplicate observations were shown for headspace CO2 and CH4 to demonstrate the variation. For organic acids, Fe(II), and pH, the standard deviations are too small to shown as error bars.

Comment 19: Figures: In general, would it be possible to add error/uncertainty bars to the data points in the figures? This would place the modeled sensitivity in the context of the measurement error.

Response 19: see second half of response 18.

Comment 20: P16L5-7 WHAM is an aqueous pool model, claiming that there is no

needed modifications when applying it to a well structured soil column seems a bit of a stretch.

Response 20: Windermere Humic Aqueous Model (WHAM) appears to be an aqueous model from its name. In fact, it treats the binding sites in soil organic matter as surface sites, and include other minerals such as Fe(OH)3, Al(OH)3, etc. In our implementation, we use surface sites to simulate organic matter. Namely, it does include solid phase, which is critical for soils.

Comment 22: Table 2 Formatting needs to be fixed for the table entries and I would suggest replacing TOTC with Total Organic Carbon and WEOC with Water Extractable Organic Carbon. OC is a common enough abbr. that it could be used here without explanation but TOTC and WEOC are not.

Response 22: Spell out TOTC and WEOC.

Comment 23: Figure and Table captions: Figure and table captions need to be able to stand alone in the manuscript, people will often scan the figures to get a sense of the results of the manuscript. Please expand the figure captions to more fully reflect the conclusions being illustrated here, this is particularly needed for the supplemental figures.

Response 23: This is improved as shown in the marked revised version of the manuscript and supplement. Please see the enclosed documents for details.

==================== Todd-Brown, K. E. O., Randerson, J. T., Hopkins, F., Arora, V., Hajima, T., Jones, C., Shevliakova, E., Tjiputra, J., Volodin, E., Wu, T., Zhang, Q. and Allison, S. D.: Changes in soil organic carbon storage predicted by Earth system models during the 21st century, Biogeosciences, 11(8), 2341-2356, 10.5194/bg-11-2341-2014, 2014. Wieder, W. R., Allison, S. D., Davidson, E. A., Georgiou, K., Hararuk, O., He, Y., Hop-kins, F., Luo, Y., Smith, M. J., Sulman, B., Todd-Brown, K., Wang, Y.-P., Xia, J. and Xu, X.: Explicitly representing soil microbial processes in Earth system models, Global Biogeochem. Cycles, 29(10), 1782-1800, 10.1002/2015GB005188, 2015. Steven D. Allison and Allison, S. D.: Brown Ground: A soil carbon analogue for the green world hypothesis?, Am. Nat., 167(5), 619-627, 10.1086/503443, 2006.

The marked revised manuscript and supplement are enclosed as supplement in this response.

Please also note the supplement to this comment:
http://www.biogeosciences-discuss.net/bg-2016-207/bg-2016-207-AC3-supplement.zip

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