Biogeosciences Discuss.,
https://doi.org/10.5194/bg-2020-222-RC1, 2020

Biogeosciences Discussions

Responses to (in italic font):

Interactive comment on "Representing methane emissions from wet tropical forest soils using microbial functional groups constrained by soil diffusivity" *by* Debjani Sihi et al.

Anonymous Referee #1

Received and published: 22 August 2020

The study "Representing methane emissions from wet tropical forest soils using microbial functional groups constrained by soil diffusivity" by Sihi et al. tries to explain soil methane emission dynamics in tropical forest soils of Puerto Rico during normal and drought conditions. They combine field measurements with modelling efforts (Microbial Model for Methane Dynamics-Dual Arrhenius and Michaelis Menten (M3D-DAMM). Overall, I think it is a really nice study that tries to combine microbial with biogeochemical data to investigate ecosystem methane dynamics. However, I have some general and some minor comments.
Thank you kindly for the positive comments and for the constructive suggestions.

The authors should describe the concept of "microsites" more in detail. The authors focused on the top 10 or 10-30 cm of their Ultisol although methane production/consumption dynamics in the deeper clay-accumulating horizons may be more important for the overall net methane emissions from their Ultisols. The authors do not discuss that and do not compare with other soils. How does the abundance of microsites change with soil type, soil depth and other ancillary variables?
Our sampling strategy, both of soils and of soil water, are geared to accompany the greenhouse gas flux measurements which are taken on the soil surface. Past studies (Silver et al. 1999) have taken methane concentrations at depth in similar soils, and found that concentrations are higher at 10 versus 35 cm. Examination of the soils in the Luquillo mountains have found that SOC maximums are around 35 cm depth (Johnson et al. 2014). For this site, clay is abundant at all depths, e.g., 20-30% clay at 0 to 10 cm depth (L101). Therefore, we believe that our sampling strategy is appropriate for the mechanisms we are trying to address. We will add this information and the Johnson citation to a revision to ensure readers also understand this concurrence of sampling strategy and past observations.

We will address the issue of comparison to other soils in response to a comment below.

There are no specific measurements of microsites at any depth at this site; microsites are inferred because of decades of observations of co-occurrences of oxygen concentrations in the soil and methane fluxes; and because of the rich clay, iron oxides, and visible redox mottling, particularly evident in the valley and slope soils (papers cited in L66-74). Techniques for measuring microsite activities remain very limited to date, here or elsewhere. We will add this note to this effect in a revision.

Minor comments: L22-25: Is it important to give this information? I would only include the most significant ones that support your guiding questions! What is the difference between "<" and "«". The abstract should self-explanatory.
We will simplify the abstract and the manuscript throughout by using only <, =, and >; it is perhaps a subjective difference between "<" and "<<". The geochemical parameters in L22-25 were measured and are key model inputs, so we will rephrase to say they were measured at the field site, but we will remove the parenthetical material regarding differences in their values with respect to topographic positions which is not germane to the abstract.

L33: write aceticlastic methanogenesis instead of “acetotrophic” and “acetoclastic” (check the whole manuscript)
Thank you for catching these typos; we will search the entire manuscript to ensure they are always correct. I am confused as to whether it is “aceticlastic” or “acetoclastic” methanogenesis, as I see both in the literature. We will clarify which is appropriate, perhaps with Biogeosciences Editorial Staff, and use it consistently in the revision.

L43: what are wet tropical forest soils? What are wet tropical forests and what is the difference between wet tropical forest soils and upland soils? How do you define that?
The soils are classified as wet tropical forest soils according to Holdridge life zones, which considers rainfall, elevation, latitude, humidity, and evapotranspiration, as described specifically regarding the Luquillo Experimental Forest (Harris et al. 2012). Upland refers only to topography. The soils in our current study (including the valley soils) could be referred to as “upland” in that they are all located in a lower montane region (~350 m elevation). We don’t believe any changes to MS are warranted in response to this comment.

L53-65: What are the soil types in the different studies? Since methanogenesis and methanotrophy are substrate-limited, the soil type with its specific biogeochemistry is very important. The authors mention that there are several studies that report effects of drought on net methane emissions across different wet tropical forest soils. Consequently, they should mention the different soil types.
We will add the soil type (below) when the studies are mentioned in L53-65 and elsewhere as relevant.
Aronson et al. 2019 (Costa Rica) Oxisols
Davidson et al. 2004, 2008 (Brazil) Oxisols
Wood and Silver 2012 (Puerto Rico) Ultisols with a similar climate and parent material
O’Connell et al. 2018 (Puerto Rico) Ultisols, same soils as current study

L66-74: Oxygen may not be the only factor for methane emissions upon rewetting. The observed rapid flush of methane in response to a wetting event may be driven by rapid depletion of other electron acceptors, as well. The major focus of this paragraph is on oxygen but what is with acetate, H2 and CO2?
We concur with the reviewer’s suggestions; in fact, that is part of why we initiated this study. We wanted additional data on acetate concentrations, and to use a predictive model to better understand the conflicting/collaborating roles of oxygen, substrate, and microbial biomass in controlling methane emissions. In a revision, we will clarify the role of these different substrates in controlling methane emissions in this paragraph so it does not appear we are only focused on oxygen. Although they are important, acetate and particularly H_2 measurements are seldom available.

L85-86: and why not H2 and CO2? How do you account for acetate formation during fermentation and homoacetogenesis? How do you account for syntrophic acetate oxidation? How can you explain the “contrasting patterns of observed CH4 emissions”, when sources and sinks of acetate etc. is not measured or simply not known?
The idea of this paper is to consider a relatively simple set of mechanisms and see how well these mechanisms can explain the complex observations. This study used a model where the production of methane is modeled by acetoclastic and hydrogenotrophic mechanisms only, and consumption by methanotrophy. Acetate and CO2 are inputs based on measurements of soil water and pH. Acetate is formed by fermentation and by homoacetogenesis as defined in Xu et al. (2015) in Eq A15 and 16 in the appendix, see also revised Fig. 1b. The model is not completely comprehensive and syntrophic acetate oxidation is neglected. In modeling, it is important to balance parsimony and mechanisms; we have made choices here to avoid overfitting. We describe the impact of some neglected processes in Section 4.3. We will add that homoacetogenesis is included and that syntrophic acetate oxidation is neglected in the methods Section 2.4.1 in a revision.

L97-105: The dominant soil type in the current study is Ultisol. I guess the whole

methane cycling will be very different in other tropical soils (What is with Oxisols?) What are the soil types in the other studies mentioned in the introduction? It would be great to see a more detailed description of the soil type. How did bulk density change with depth? At what depth does clay accumulation (subsurface zone) start? How big is the eluvial horizon?
The response to this comment also includes response to the first detailed review comment (comparison to other soils). The soil types of the other tropical studies of methane releases from the introduction are mentioned in a reply above. The studies in Costa Rica and Brazil were specific in showing that methane consumption was the major effect of the seasonal El Niño cycle (Costa Rica) or imposed drought manipulation (Brazil). Only the O'Connell et al. (2018) study showed the enhanced release of methane during post-drought recovery. We will add this distinction clearly in a revision. We cannot say if the O'Connell observations are related to soil type or some other mechanism. Oxisols, because of their high oxide and clay contents, may also have microsites.

We have data on bulk density changes up to 1 m that are in review with Ecology and Evolution. In that manuscript we find that on the surface, bulk density ranges from 0.5 to 0.7 g/cm3. By 25 cm depth, bulk density is 0.8 to 1.1 g/cm2. These results are similar to those published by Johnson et al. (2014).

A publication that provides a detailed soil survey in the immediate vicinity of the field site (Soil Survey Staff, 1995) lists the following soils: Zarzal, Cristol, and Prieto soils. For all soils, the litter layer is minimal. For the Zarzal soil, the surface (A) horizon is usually 4 cm thick, and the B horizon is around 150 cm thick. The Cristol soil surface horizon is listed as 6 cm thick, and B horizon to around 150 cm thick. The Prieto soil surface horizon is listed as 6 cm thick, and B horizon to around 130 cm thick.

L107: Why was soil only sampled from the top 10 cm of the soil? I guess methane consumption dominates at the surface due to oxic conditions while in the deeper soil horizons methane production dominates due to more anoxic conditions.
The soil flux chambers are on the soil surface and the sampling strategy was designed to focus on near-surface measurements, as described above and in another comment response below. These soils are very rich in clays, beginning at the interface with the atmosphere, and are often very wet, and decades of lab- and field-scale studies by the Silver group (see various citations in the manuscript) confirm that methane production can occur in soils at shallow depths, especially in the valley soils but to a lesser extent in the slope and ridgetop soils.

L111-114: What was the detection limit for acetate?
We assumed it was equivalent to the lowest standard by HPLC analysis, i.e., 0.5 μM.

L117-120: The chemical data (what chemical data? Only pH?) for bulk soil is from the 0-10 cm soil depth and acetate and DOC from the pore water from 10-30 cm soil depth? I do not understand how you relate this information, taken from different soil depths, to each other. Finally, where do you find or where do you assume these microsites? (only in the top 10 cm or below? Or may be even below 30 cm?
We assume that soil flux chambers placed on the top of the soil surfaces are dominated by fluxes from relatively shallow depths. This summary of fluxchamber methods by two experts in this field states that it is usually assumed surface chambers measure fluxes from about 25 cm in depth (Rochette and Hutchinson 2005). Given this perspective, we collected soil and soil water measurements from the 0 to 30 cm depth to best relate to surface flux chamber measurements. The chemical data used in this study consisted of acetate from the lysimeters located at a maximum depth of 10 cm and a maximum depth of 30 cm. The soil samples on which pH was measured and from which DOC was extracted were collected from 0-10 cm depth.

Microsites are inferred by observations such as originally presented in O'Connell et al. (2018), i.e., sudden releases of methane during post-drought recovery; and from seminal publications such as Silver et al. (1999). In the latter, co-occurrences of soil oxygen and methane in bulk soils presume the abundance of anaerobic microsites in these upland soils. Because the soils have abundant clay and iron oxides at all depths, it is likely that microsites are pervasive throughout.

L144-148: What is with methylotrophic methanogenesis. You should discuss about the potential contribution of methylotrophic methanogenesis (see Norrow et al. 2019).

Our model does not include methylotrophic methanogenesis. We will add this to the mention of several other processes not considered in our model in Section 2.4.1 and cite the relevant paper.

L173-174: Why not?

We believe the reviewer is asking why iron reduction and oxidation are not included in this study. As explained in Section 4.3, we chose to take a more simplified approach to start, just focusing on substrates and microbial functional groups for methanogenesis (both acetoclastic and hydrogenotrophic methanogenesis) and methanotrophy. We feel that our model does a decent job at reproducing the data, considering "normal" and "drought" conditions, and involving two different time frames of data collection, and we consider that as confirmation of the validity of our approach. We acknowledged in Section 4.3 that iron reduction can alter the pH of the soils and soil water and enhance methane emissions; and that iron reduction can also support anaerobic methane oxidation, as well as using acetate as a substrate and thereby reducing net methane emissions. We acknowledge that our model fits are not perfect, as you can see from Fig 3, the model misses the highest methane fluxes seen during the post drought. This could be a result of not considering the pH effects of iron reduction that enhance methanogenesis. However, other processes in the iron cycle reduce methanogenesis, so the benefit of including unconstrained iron cycling processes is unclear without additional information to constrain the model. Therefore, we felt it was appropriate to focus only on the mechanisms covered in this study.

L206: Why 15 cm?

Please refer to the response for comments regarding L117-120 (above).

L214: How do calculate "total microsites"? I would assume that there are way more microsites in clayey horizons below 15 or even 30 cm soil depth? Overall, I think you have to explain the concept of microsites, more in detail? You are scratching only the soil surface at the moment but in my opinion the biggest methane production potential occurs in deeper parts of the Ultisol.

A seminal study in 1999 by Silver et al (cited in our manuscript) in a nearby Ultisol soil took measurements of methane and oxygen at 10 and 35 cm depth, as well as surface chamber flux measurements. The authors found that methane concentrations were higher at the shallower depths. Most of the subsequent papers from Luquillo Experimental Forest used surface chamber measurements and focused on shallow soil depths.

We assumed that size of the microsites should be at least an order magnitude lower than the bulk soil measurements we had for soil methane fluxes. Using this logic, we decided that "diameter" of microsites should be in "mm" scale as the diameter of soil chambers we used are in "cm" scale (15.24 cm). Thus, we did the math to come up with the number of "total microsites" (i.e. 10000) such that the diameter of microsites meets our criteria. We will add this to methods Section 2.4.2.

L270-272: Why are acetate and hydrogen production decreasing when aceticlastic and hydrogenotrophic methanogenesis also decrease? Aceticlastic and hydrogenotrophic methanogens consume acetate and hydrogen, respectively. So, if there is a decrease in aceticlastic methanogens, I would first assume an increase in acetate concentration and thereafter a sharp decrease if oxygen levels further increase.

Decreasing acetate and hydrogen production in the model are consistent with decreasing acetoclastic and hydrogenotrophic methanogenesis. Eq 1, biomass amounts and substrate concentrations together contribute to the reaction producing methanogenesis; it is not sequential.

L280: How do explain that?

We believe the reviewer is referring to predicted changes in the biomass of different microbial functional groups during drought, drought recovery, and post-drought. These are model predictions, that are based upon the mechanisms within the model, and the input data that constrains model behavior (pH, acetate, DOC, and CH_4 fluxes). Although we lack measurements of the microbial biomass of specific microbial taxonomies or functional groups during the events in this paper (as we have acknowledged), microbes can respond rapidly to changes in their environment. It is important to distinguish that the model is predicting changes in biomass of a particular microbial functional group and is not predicting large changes of the bulk microbial biomass in the soil. Bulk microbial biomass in the soil is likely to double or perhaps quadruple in response to changes in conditions, but individuals can grow

exponentially (Goberna et al. 2010; Pavlov and Ehrenberg 2013; Roussel et al. 2015; Buan 2018).

L286-287: Why does the increasing production of acetate lowers the pH?
Acetate production is a source of proton (L375 and citations therein, particularly Xu et al. 2015) as seen in Eq 7 and Fig. 1.

L301-306: If the diffusion of H2 increases during drought, one may think that hydrogenotrophic methanogenesis should increase as well. However, it does not increase because of increasing oxygen levels. That should be made clear!
This is true, particularly for the ridge and slope soils (Fig. S6), but is less true for the valley soils because gas diffusion remains limited throughout the event in the valley soil (Fig. S8). This explanation is also consistent with the methanotrophic biomass (Fig. S5). We will revise accordingly.

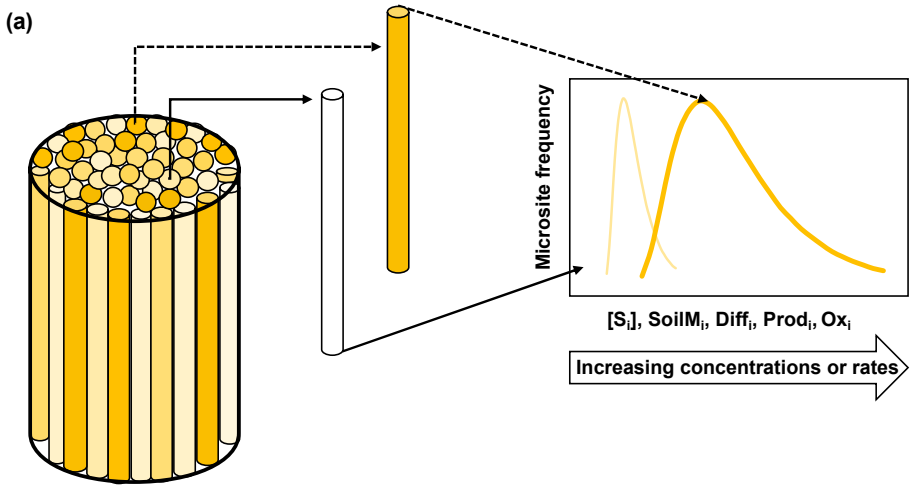
L304-309: The diffusion of acetate increases upon rewetting but that of H2 decreases. Why do you observe an increase in overall gross methane production. First, I would assume that under relatively acid conditions, hydrogenotrophic methanogenesis dominates. I think the overall increase of methane emissions upon rewetting is because of oxygen depletion and therefore the stimulation of methanogenesis in general . . .. and not because of increasing acetate concentrations or a shift in the methanogenic pathway of methane formation. If it is really aceticlastic methanogenesis that is stimulated, you should provide some isotopic data. There is competition for acetate between several microorganisms. In the end it could be simply stimulation or inhibition of fermentation or homoacetogenesis that drives changes in the amount of observed acetate concentrations.
In the model and likely in reality, all of these processes are occurring simultaneously, although some may be more important than others. The purpose of this paper is to better understand the unique processes observed in the 2015 drought, that do not have parallels in either Costa Rica or Brazil. A model was used that was originally validated by simulating methane emissions in Arctic soils (Xu et al. 2015), but the experiments were lab-scale incubations. Therefore, the model was enhanced here to consider diffusional processes that may be important at the field scale. From these beginnings, the model seems to explain the observations from the 2015 Puerto Rico drought, and 2016 "normal" scenario. It is to some extent a thought exercise, i.e., "If we were to include the processes X, Y, and Z, could we reproduce the observed data?". If the answer is yes, then that indicates the model provides valid explanations for the observations. However, we acknowledge that the model is not fully comprehensive – in the interest of parsimony, some processes are excluded. And that the presentations in the paper are model simulations, and that in some cases, true validation data, e.g., microbial biomass of the different functional groups or H_2 concentrations or isotopic data, is lacking. We believe this exercise is valuable, despite the shortcomings therein, and we have tried to be open about where either measurements or model processes are lacking.
The mechanism of hydrogenotrophic methanogenesis does not well explain the observations. Hydrogen should have been freely available during the drought, yet little methanogenesis was observed. As the soils wetted, hydrogen would become less available as its diffusion rate will decrease strongly (Fig. S8). So hydrogenotrophic methanogenesis cannot readily explain the post-drought spike in methane concentrations. Acetate diffusion, however, more readily explains the observations (Fig. S8). As wetting commenced and progressed, acetate may become more available to microorganisms and can enhance methanogenesis.
At the same time, wetting decreases oxygen availability, decreasing the role of methanotrophy (Fig. S8) and allowing more methane to escape the subsurface, despite limitations in gas diffusion.

L318: Again, how do you define the microsites?
We are sorry that microsites are inadequately defined in this version. We have revised the figure and caption as follows:
"Top panel (a) shows the model representation of soil microsite distribution (modified from Sihi et al., 2020a, also see Eq. 14). The cylinder refers to the volume beneath the soil chambers. The intensity of different cylinder colors figure refers to rate of a process or the intensity of a concentration inside microsites in each theoretical cylinder, e.g., a dark color means a higher rate/intensity, and a light color means a lower rate/intensity for a given process. The 2D graph on the right refers to the probability density function of the rate of the process or intensity of the concentration in the bulk soil. A wide distribution skewed to the right (dark-colored line) implies higher bulk rates of the

process or higher concentrations, and a narrow distribution skewed to the left (light-colored line) implies lower bulk rates of the process or lower concentrations, of any of the following: soil moisture, solute concentration, gas concentration, gas diffusion, solute diffusion, methane production, or methane oxidation."

FYI, we have revised the figure below from the original as follows: Added an arrow on the x axis pointing towards the right, denoting increasing concentrations or rates. Moved the light-colored function to the left of the dark-colored function and made it much more narrow and signify less impact on bulk rates/concentrations compared to the dark-colored function (more impact on bulk rates/concentrations).

Please see revised Figure below:



L346-348: Again, what makes you so sure that it is acetate that drives net methane emissions and not H2/CO2 and a decrease in oxygen?
All of these processes are happening simultaneously. Our model simulation suggests that acetate is driving most of the methane increases, and that decreases in methanotrophy due to decreases in oxygen, are both more important than hydrogenotrophic methanogenesis, pls see also sensitivity analysis in Fig 8. Fig S5 shows that both kinds of methanogens increase during drought recovery and post-drought, but that acetotrophic methanogens are two orders of magnitude more abundant than hydrogenotrophic methanogens. Additionally, the acetate hypothesis makes more sense because under drought conditions, acetate may accumulate in microsites. During wetting, the acetate may become more available to the methanogens as solute diffusion becomes enhanced (Fig. S8), resulting in strong methane releases. Hydrogen, on the other hand, would be readily available during drought conditions because its diffusion would not be limited. So, hydrogen substrate availability does not explain the observations of strong methane releases under wetting conditions. In fact, the model simulations suggest that hydrogen diffusion is lessened under wetting conditions (Fig. S8).

L369-372: and homoacetogenesis?
Yes. We will specify in Methods Section 2.4.1 and in Fig. 1b caption that this mechanism is included in the model.

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Interactive comment on Biogeosciences Discuss., https://doi.org/10.5194/bg-2020-222, 2020.