Overall response

First, we thank Reviewer #2 for taking his/her time to read through our paper. However, we are disappointed that Reviewer #2 largely misunderstood our development and analyses, while Reviewer #1 has grasped the important concepts and agrees with their relevance and importance. Below we respond to Reviewer #2's comments. However, given Reviewer #2's (1) large number of misunderstandings of our work; (2) fundamentally incorrect criticisms, and (3) poorly constructed suggestions for improving the manuscript, we request an alternative reviewer to evaluate our paper and responses.

Among other problems described below, Reviewer #2 misclassified our work into the category of ecological "aggregation" of "micro-dynamics" into "macro-dynamics", a topic well trodden by previous researchers in ecology. We are very aware of the studies mentioned without citation by the reviewer. Briefly, the so-called "aggregation" approach, as studied in ecology (and also in theoretical economics), is a mathematical technique of dimensional reduction. This approach assumes that the micro-dynamics is available for "aggregation" so that the resultant macro-dynamics retains as much of the micro-dynamical functional responses as possible. Such work falls broadly in the category of "reduced order modeling", a topic on which we have recently published a number of papers (e.g., Liu et al., 2016; Pau et al., 2014, 2016). In stark contrast, the study we present here describes an approach to formulate the micro-dynamics in a physically consistent manner. Therefore, the reviewer's primary summary criticisms are irrelevant to our study. Further, the reviewer's comments contain several blatant errors, which we detail below.

Comment: In this paper, the authors go through many different formulations of enzyme kinetics in an attempt to 'scale' kinetics from a single enzyme system to a metabolic network consisting of 10's to 100's of reactions. The paper doesn't have a good introduction and there is little motivation for why it's so critical to be able to 'scale' enzyme reaction kinetics other than it's computationally intensive to simulation a bunch of equations instead of one. Reducing dimensionality will always make life easier, but it's not clear that anyone in a real world modeling situation would even be in a position to try to translate kinetics for 10's to 100's of reactions for soil organic matter decomposition to reduced set of reactions. In a real network, there will be feedbacks between reactions but all are considered independent in this manuscript.

Response: Given reasonable space constraints for journal articles, we did not detail the huge literature on enzyme kinetics relevant to soil biogeochemistry. However, we included sufficient references (Allison, 2012; Bouskill et al., 2012, Grant et al., 2016; Riley et al., 2014; Sulman et al., 2014; Tang, 2015; Tang and Riley, 2013, 2015; Wieder, 2013, 2014) on the topic to indicate that soil biogeochemical models are now in a position to include a wide range of biogeochemical reactions, which may very well exceed 100 reactions. For instance, the soil biogeochemistry module of the site- to regional-scale ecosystem model *ecosys* (e.g., Grant et al., 2015, 2016; Mekonnen et al. 2016) represents a wide range of microbes, including heterotrophic aerobic bacteria and fungi, methanogens, methanotrophs, autotrophic ammonia oxidizers, autotrophic nitrate oxidizers, acetogen fermenters, and autotrophic and heterotrophic nitrogen fixers. The model also represents the aqueous chemistry of phosphorus dynamics that involves iron, calcium, carbonate, etc.

We also never stated that our goal was to reduce the reaction network. Rather, we emphasized the need for formulation consistency between the many reactions in describing the substrate-consumer relationship, which is the first step in modeling soil biogeochemistry (as indicated in the title, abstract P1: L12-13, and throughout the main text). If formulation reductions were proposed, they should only be applied to substitutable substrates (as discussed in section 3 and also in Tang and Riley, 2013). For example, aerobic heterotrophic bacteria can feed on proteins, cellulose, carbohydrates, and starch; if the specific evolution of those chemical compounds is not of interest, we can regard them all as carbon substrates, which is the fundamental assumption that has been widely applied in the development of many soil BGC models (e.g., RothC model (Coleman and Jenkinson, 1996), CENTURY model (Parton et al., 1998)). A similar problem involving enzyme interactions with many substrates was also studied in Schnell and Mendoza (2000). Given soil microbes are competing and collaborating with each other to consume the many chemical substrates, our study is definitely relevant to modeling complex soil BGC networks. We also acknowledged that feedbacks between reactions are critical components of the soil BGC network (e.g., on P4: L13-17, we acknowledged that there are temporal and spatial scaling methods to cover those feedbacks). Overall, our formulation attempts to better resolve the interactions and feedbacks between reactions at the microbial uptake stage (i.e., the consumer-substrate interactions), which is a misunderstood or ignored topic in the literature (see the long review in Tang and Riley, 2013).

Therefore, this reviewer's comment both misses the point of our manuscript and mischaracterizes its relevance. To ensure readers who're unfamiliar with soil biogeochemical modeling not to confuse our study with dimension reduction through so called "aggregation", we added a new paragraph in page 4 in the revision to state specifically that we're attempting to improve the microdynamics.

Comment: The authors quickly jump into kinetic equation after equation with no clear goal and minimal to non-existant links between models/equations.
Response: These comments are somewhat shocking given the manuscript's theoretical development goals are given in the Title, Abstract (P1: L11-15), and Introduction (P4: L6-18, P10: L13-22, P11: L1-6). Clearly, we are proposing the SUPECA kinetics to (1) scale redox reactions in networks of mixed substrates and consumers; (2) consistently address the interactions between substrates and microbes at the substrate uptake stage in modeling soil biogeochemistry; and (3) demonstrate its applicability using a simple aerobic soil respiration problem.

Comment: The authors never even clearly articulate why what they are presenting is better than anything else. The manuscript is incredibly hard to follow as well. It may be possible for the authors to distill some of this down into a coherent compelling message, but in its current form it's not publishable.
Response: This comment is again strange, given our substantial discussion in the Abstract (P1: L15-23, P2: L1-11), Introduction (P5-10), and sections 3, 4, and 5. Throughout our discussion, we also highlighted problems with the current formulations of soil BGC kinetics.

For the editor and reviewer's information, and to put the value of this work in

context (i.e., "why it may be better than anything else"), the formulation we described in this paper follows the work described in Tang and Riley (2013), where we originally described the Equilibrium Chemistry Approximation. In this context, we note that Jinyun Tang received the Ecological Society of America's Honorable Mention for the Gene E. Likens Award for this paper (indicating it, at least, may have value compared to other approaches). Further, the ECA concepts are actively being applied in site to global-scale modeling efforts (Zhu and Riley 2015; Zhu et al., 2016a,b; 2017), which we cite in the manuscript. We therefore believe these reviewer's comments indicate a misunderstanding of our paper and the broader modern literature on numerical model representations of biogeochemical processes.

Comments: Pg. 7, line 12: This doesn't make sense. The whole idea is to consider a network of interactions, each with their own kinetics. Gardner, O'Neill, and Iwasa, among other did seminal work on aggregating model dynamics and establish good rules of thumb for when aggregation is reasonable. The problem the authors of this manuscript are trying to address is one of aggregation, not scaling. Furthermore, their expressions are incorrect. A sum can be expressed as the number of terms in the sum multiplied by the mean of the sum. In their case, each term is a product of a rate constant and a concentration, which means that impossible to make their substitution. At a given instant, it can work, but a soon as concentrations change their expression is invalid.
Response: This comment is again a misreading of our work. First, we are addressing the substrate-consumer relationship, an important component in formulating the microdynamics; whereas the works by Gardner, O'Neill, Iwasa (see our listed reference on aggregation) and others are on aggregating the microdynamics when the latter is given. Second, we did not indicate that we are averaging nonlinear terms. Even when we sum (and average) the terms in equation (7), we state clearly that the kinetic parameters must be equal for such a summation (P9: L6). Throughout the paper, we used the summation and average rules according to standard practices widely used in mathematics and physics, and therefore this criticism appears baseless.

Comments: Furthermore, there's no way to average the nonlinear interaction between enzyme and substrate for multiple reactions. I tried looking up the partition principle and didn't find anything, and the analogies with Dalton's and Newton's laws don't make any sense.
Response: First, we did not contend that we are averaging the nonlinear interactions between enzymes and substrates for multiple reactions. Second, we introduced and defined the "partition principle" on page 6, line 16. The concept is widely used in deriving macroscopic representations of complex phenomena in physics (e.g., Dalton's law of partial pressures; superposition principle of electrostatic forces, angular moment etc.; Feynman et al, 1963), and we argue in this manuscript that it should be applied in developing representations of soil BGC dynamics. Third, we only apply averaging when the relationship is linear and there is a good conceptual understanding to support it (e.g., equation (7)). For instance, as we explained above, some models of soil organic matter decomposition aggregate different organic matter constituents (e.g., protein, cellulose, carbohydrates) into a single carbon pool, and still provide important scientific insights to the soil carbon cycle.

Comment: Pg. 3, line 23: Wieder, not wider
Response: Thanks for pointing this out. We corrected it in the revision.

Comment: Pg. 6, line 2: dissociation
Response: Thanks for pointing this out. We corrected it in the revision.

Comment: Pg. 6 line 7: r-K selection is only briefly mentioned in the Klausmeier and Litchman (2008) paper.
Response: Yes, we agree, so we added a citation to Tilman's work (Tilman, 1982) for readers interested in this topic.

Comment: Pg. 7, eq. 2: Both terms are negative but dissociation should be positive
Response: We think there is a misreading of equation 2. Only the first term is negative, the second term is positive and describes dissociation.

Comment: Pg. 8, line 1: I have no idea what the nonsingularity principle is, and again, searching for it gave no results. The expression is really conservation of mass anyway.
Response: We apologize that we did not originally provide a citation for this concept on page 8 (it is mentioned on Page 9, lines 1-3 and the singularity is defined in P2: L1-L3); we have now added references on the concept at the first appearance of the term in the revision (Schnell and Maini, 2000; Tang and Riley, 2013; Tang, 2015).

Comment: Pg. 8, line 12: I don't know what this means. Furthermore, the only difference between the two sides of the equation is that the r.h.s. just moves the half saturation constant around. They appear equal and there is no basis for why they wouldn't be.
Response: No, in equation (6) the term after the first equal sign is not equal to the term after the second equal sign, as we show below:

Suppose there are two substrates, S1 and S2, with concentrations of 1 and 2 units, respectively; and half saturation constants of 1 and 2, respectively. Then, assuming all other parameters are of numerical value 1, the value after the first equal sign is 1/(1+1)+2/(2+2)=1. However, the value after the second equal sign is (1/1+2/2)/(1+(1/1+2/2))=2/3. Therefore, they are not equal (i.e., $1 \neq 2/3$). Such a case will occur, for instance, in situations when both NH_3 and NO_3^- are taken up by a microbe or plant to synthesize biomass. Only the term after the second equal sign will describe this uptake process consistently. A similar situation is discussed in detail in Schnell and Mendoza (2000).

Comment: Litchman and Klausmeier (2008) don't even mention Monod kinetics. It is unacceptable to incorrectly use references to justify assumptions or manipulations.
Response: It seems the reviewer misunderstood the reference to Litchman and Klausmeier (2008). In their page 620, the second equation, which we copy below, is the Monod kinetics:

$$uptake = v(R) = \frac{v_{\max} R}{K + R}$$

with R is the substrate and K is the half saturation constant.

Even though Litchman and Klausmeier (2008) did not use the term "Monod kinetics", they are clearly applying that approach. As the reviewer may be aware, Monod kinetics and Michaelis-Menten (MM) kinetics were proposed based on different empirical evidences. The Monod kinetics is purely empirical (Monod, 1949) and MM kinetics can be derived mechanistically (Briggs and Haldane, 1925). In soil biogeochemical modeling, the Monod and MM kinetics are used for modeling microbial substrate uptake, and under the assumption of no substrate-storage in microbial cells (which is valid under some restrictive conditions), the Monod kinetics and MM kinetics (or any substrate kinetics such as the SUPECA we present here) can reasonably represent microbial growth (Monod, 1949; Wieder et al., 2013, 2014; Tang and Riley, 2015).

Comment: Pg. 9, line 3: When is it even reasonable to enzyme concentration approach infinity?
Response: It is a common practice in deriving macroscopic representations of complex phenomena to ensure that the solutions are robust across a range of conditions. The term "approach infinity" is widely used in scientific literature to imply "as a state becomes large compared to another state" (e.g., see chapters on oscillators and electrostatics in Feynman et al., 1963). In biogeochemistry, for example, such a situation exists in vivo conditions inside an organisms' cell (e.g., Schnell and Maini, 2000), or when mineral surface interactions are represented analogously to enzyme kinetics (e.g., adsorption is of Langmuir type). In such situations, the ratio of enzyme and substrate concentrations becomes very large (i.e., approaches infinity).

Comment: Pg. 9, line 5: There is no paper that I can find that matches the Murdock reference, and one published in the same year is completely unrelated. This is a very disturbing pattern of misrepresentation of the literature. I basically can't follow the rest of page 9 and I have no idea what parametric sensitivity is.
Response: It appears the reviewer was searching for a citation by "Murdock", when the paper we cited is by "Murdoch". The reviewer's assertion of a "disturbing pattern of misrepresentation of the literature" is ridiculous and unprofessional, considering that he/she could have simply gone to the reference list at the end of our manuscript and found the citation.

In 1973, William W. Murdoch published two papers, one is "The functional response of predators", and the other is "Predation by Coccinellid Beetles: Experiments on Switching" which he co-authored with J.R. Marks. The first paper is the one we cited (and is listed in the references).

The term "parametric sensitivity" is a widely used term in numerical modeling, and we cited a recent paper on the topic in the original manuscript. However, there are many other recent publications applying this term; we have therefore added some of those citations (e.g., Qian et al., 2015; van Werkhoven et al., 2009).

Comment: Starting in section two, the 'derivations' seem to be ok, but they are trivial algebra. It's easy to start with any reaction diagram, assume quasi steady state and derive equations. However, they still seem to retaining more dynamics that is typical because the substrates/reactants A and B are changing over time.

Response: Our careful derivation attempts to present nuances to readers, and indicate clearly where critical assumptions are being made. Since one of our clearly stated goals is to formulate a consistent set of reaction kinetics for soil BGC, we believe having a consistent derivation formulated in the peer-reviewed literature is important. Further, we used this derivation to describe possible problems with other approaches in characterizing biogeochemical kinetics, such as dual-Monod kinetics and synthesizing unit kinetics. Therefore, our derivation will help readers to understand the uncertainties behind using those kinetic formulations for their modeling analyses. In the same spirit, throughout the paper, we have clearly reported that our new approach is only a better approximation to the law of mass action (e.g., section 4 and also see Tang, 2015), and should not be regarded as accurate for all conditions (a situation that is discussed in detail by Pedersen et al., 2008, which we have cited in the revision).

Comment: Pg. 17, lines 2-3: I have no idea what this sentence means.
Response: The phrase "MM kinetics ignores the mass balance constraint of substrate" simply means: in the derivation of MM kinetics, no constraint is placed on the substrate mass balance. Tang (2015) described this condition and discussed its implications, as have others (Borghans et al., 1996; Tang and Riley, 2013; Maggi and Riley, 2015). We have added these other citations to the revised manuscript to buttress this point. However, as the subsequent sentences explain, our point is that a similar problem may be happening in Dual Monod and Synthesizing Unit kinetics formulations.

Comment: Pg. 17, line 15 (and appendix): I have no idea what their 'first order closure approach' is. The appendix isn't really a help here.
Response: In the revision, we added some explanation to the "first order closure approach" and a citation to Tang and Riley (2013) where the approach was first applied to enzymatic chemical kinetics. We also note that the first order closure approach has been applied in many other fields, and have added citations (Shankar, 1994; Tang et al., 2007) to the revised manuscript.

Comment: Pg. 19: The problem with trying to average over a bunch of nonlinear interactions seems to render this derivation incorrect.
Response: As we discussed above, we are not trying to "average over a bunch of nonlinear interactions". Given that Reviewer #1 understood this important point, we are at something of a loss to address Reviewer #2's misunderstanding. Nowhere in the manuscript did we state we are "trying to average over a bunch of nonlinear interactions", so it is not clear where he/she developed that perception.

Comment: Data examples and figures: By this point, I am totally lost and quite skeptical of whether their derivations are correct. The comparisons with data are poorly motivated and described so it's not possible to even know what we should be taking away from the exercise and why.
Response: Although we appreciate the reviewer's taking his/her time to read through our manuscript, we believe that this reviewer is an inappropriate choice, given his/her (1) large number of misunderstandings, (2) fundamentally incorrect criticisms, and (3) poorly constructed suggestions for improving the manuscript. In other contexts we would be

happy to discuss the details of the approach with the reviewer, but given that this is a manuscript review, we request the editor to find another reviewer who's more familiar with biogeochemistry and approaches to develop conceptual and numerical models of complex reaction networks.

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Works on aggregation

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