

## Responses to the reviewer's comments:

We thank the three referees for their critical though constructive review and their valuable suggestions to improve our manuscript. To the best of our knowledge we have modified the manuscript according to all suggestions made by the referees. Below we answer each comment individually.

### Anonymous Referee #1

#### GENERAL COMMENTS

Despite the fact that tropical forests are thought to be the largest natural sources of N<sub>2</sub>O and NO<sub>x</sub> globally, we have an incomplete understanding of the magnitude and range of N<sub>2</sub>O and NO<sub>x</sub> fluxes across different tropical habitats. In particular, upland/montane ecosystems are particularly under-represented in process-based studies, bottom-up emissions inventories and modelling studies. This study is therefore interesting because it seeks to model and extrapolate N-trace gas fluxes from upland tropical ecosystems in Africa where we know little about biosphere-atmosphere exchange or ecosystem N dynamics.

However, despite the great promise of this paper, it suffers from a few critical problems. First and foremost, the modelling dataset has not been adequately validated against field flux measurements. While the investigators have attempted to parameterise their model using laboratory-based incubation data, this alone is not sufficient, given the scaling issues associated with trying to link laboratory- to field-based measurements. While laboratory measures may be sufficient to calibrate response curves of N<sub>2</sub>O and NO<sub>x</sub> flux against soil moisture/WFPS (as described in the lead author's 2012 paper), it is often unclear - a priori - if laboratory incubation data will provide an accurate estimate of the mean, median and variance/range of field fluxes without direct measurements to back them up. Another problem with laboratory measurements is that they do not always account for switches and lags in biogeochemical processes, associated with changing weather, soil or other environmental conditions. A combined laboratory/field measurement approach is often required in this kind of context to ensure that the lab and field measurements are in good agreement. While I fully appreciate that it can be difficult to collect field data in some tropical locations, at the very least if the investigators were able to conduct at least a few campaign-based measurements to validate/ground-truth their lab and modelling data, this would greatly help the manuscript and study as a whole.

*Response: We agree that our validation of the simulations was based on lab incubation data and that a validation against field based flux measurements would be better. However, we are convinced that the paper still provides a very valuable and robust estimation of N<sub>2</sub>O and NO emission from poorly investigated (African) tropical forests. Below we argue why using lab incubations was the only realistic and also acceptable option for validation of the N<sub>2</sub>O and NO simulations.*

*First we initialized the model using a unique field based data set. We did NOT use the lab incubations to parameterize or calibrate the model, but only compared simulation results of N<sub>2</sub>O and NO emissions with lab measurements. Second, we referred to other studies (cited in Gharahi Ghehi et al., 2012a) whereby average N-trace gas fluxes derived from soil incubation experiments agreed well with field-derived fluxes. Moreover, "a few field-based campaigns", as suggested by the reviewer, would not solve the problem since still spatial (mainly driven by soil and above ground biomass) and temporal (mainly driven by climate,*

*e.g. wet vs. dry season) variability of NO and N<sub>2</sub>O would not have been covered. Third, we argue that a detailed spatial and temporal collection of field-derived N-trace gas fluxes is logistically not feasible for the ca. 1113km<sup>2</sup> Nyungwe forest (e.g. remote location, steep slopes, etc.). The latter would absolutely not be possible for NO as these measurements need to be done in the field via dynamic chambers and complex analytical requirements, which is less the case for N<sub>2</sub>O. In addition, this is one of the few studies that include NO simulation for tropical systems. Nevertheless, in view of this validation criticism we have stated our simulation outcome more cautiously (see below).*

*A project to carry out field based measurements has been submitted in the meanwhile, which will include hiring local staff and setting up a laser based trace gas facility in the IITA branch office in Bukavu, DR Congo.*

Second, the model results need to be stated more cautiously and with less certitude, given that they are not adequately validated against field results. This is particularly prominent in the Discussion section, where the authors make much more sweeping statements about N<sub>2</sub>O/NO<sub>x</sub> exchange than I believe that results warrant. For instance, on page 1496, lines 19-24 rather than saying with certitude that "On average the entire Nyungwe forest emits..." it would be better to couch the model findings more cautiously; for example: "On average the entire Nyungwe forest is likely to emit on the order of 439 t of N<sub>2</sub>O-N...etc."

*Response: Also in view of the previous comment we agree with the reviewer on this point and made corrections accordingly.*

Third, the authors have identified that their model is highly sensitive to variations in bulk density (BD) and pH, yet readily admit that BD is one of the key variables missing in their legacy dataset (page 1489, lines 8-11). They have indicated that they estimated BD using some kind of transfer function, but it is not immediately clear how robust this approach is for estimating BD of the soils in question. For example, did they include some kind of error propagation calculation to account for uncertainty in modelled estimates (rather than direct measures) of BD. While the authors have cited a previous publication (Gharahi Ghehi 2012b SSAJ 76:1172-1183) that discusses this method, I think - given how critical BD is to explaining the variability in their results - that they launch a more robust defence of this approach, given that there seems to be no ready substitute for empirically-derived BD numbers in my mind.

*Response: We can assure the reviewer that we estimated BD not with "some kind of transfer function", but that this was carefully done using key experts, all available soil data from Rwanda and state of the art mathematical tools including error propagation. Finally, the predicted BD data were validated against new field based BD measurements (see full details in (Gharahi Ghehi et al., 2012b). We rephrased this in the manuscript as follows: "Since soil bulk density values are largely missing in the original survey, bulk density was derived from pedo-transfer functions (PTF) specifically developed for tropical highland forest soils with errors that are acceptable considering the typical errors associated with direct field measurements of topsoil BD" line 175-179.*

Fourth, one of the key findings of this study is that the model is unable to adequately simulate 'hotspots/hot moments' in N-trace gas fluxes, as evidenced by the poor fit between measured and simulated results for very high N<sub>2</sub>O/NO<sub>x</sub> fluxes. This tends to suggest that the model as a whole is likely to underestimate N<sub>2</sub>O/NO<sub>x</sub> fluxes, as the model isn't able to capture these 'hotspots/hot moments'. The investigators need to explore this issue more deeply in the Discussion. For example, it would be useful to

know how frequently these 'hotspots/hot moments' occurred in their incubations. Can the investigators use this frequency/probability information to estimate (roughly) how much they may have underestimated N<sub>2</sub>O/NO<sub>x</sub> fluxes?

*Response: Indeed in the manuscript we mentioned that for the 31 sites the simulated mean N<sub>2</sub>O and NO emissions were generally < 15 g N ha<sup>-1</sup> d<sup>-1</sup>. Although most (> 80%) of the laboratory N<sub>2</sub>O and NO fluxes agreed well with the simulated data (which is already a great result in it self) a few sites showed measurements that were three to four times higher (Fig. 5). These high N<sub>2</sub>O and NO emission values (>15 g N ha<sup>-1</sup> d<sup>-1</sup>, Fig. 5) were indeed somewhat surprising. We did a careful assessment of the latter high NO and N<sub>2</sub>O fluxes and came to the following conclusions:*

*N<sub>2</sub>O emissions:*

*All 5 sites with mismatch of measured and simulated N<sub>2</sub>O emissions have highest values of clay content and thus highest values of soil moisture. Climate in the Nyungwe is very wet leading to overall high values of soil moisture at field conditions. Therefore, in particular model simulations predicted also substantial N<sub>2</sub> emissions (data not shown) (again in particular for the 5 sites mentioned above). Values for these sites are about 5kg N ha<sup>-1</sup> yr<sup>-1</sup> higher which is about 15 g N ha<sup>-1</sup> d<sup>-1</sup>. If we would add this to the simulated N<sub>2</sub>O emissions, simulated and measured values would agree much better. It is also worth mentioning, that in contrast to simulations, measurements might have the problem to overestimate N<sub>2</sub>O emissions, due to the fact that the activation of N<sub>2</sub>O (and NO) reductase can be delayed. Such a delay (implemented in the model see Kiese et al., 2005) can be explained by the stepwise initialization of denitrification enzymes after the transition from aerobic (dry conditions, in our case air dried samples) to prevailing anaerobic conditions (wet conditions, in our case rewetting), which can last from hours to days (e.g. Koerner & Zumft, 1989; Baumann et al., 1996; Otte et al., 1996). The latter might be reflected by general increase of measured N<sub>2</sub>O emissions with soil moisture (even at the high level of 90% wfps) where one might expect a decrease due to increase in the N<sub>2</sub>: N<sub>2</sub>O ratio. Finally, in this respect using simulated soil moisture in 10cm soil depth likely overestimates soil moisture for a soil depth of 0-10cm (dimension of soil samples use for experiments). Furthermore, pH values of the 5 soils are rather low (mostly < 4) which can also inhibit pH sensitive N<sub>2</sub>O reductase (e.g. Kesik et al., 2006) in the measurements which might be underrepresented in the model parameterizations of denitrification.*

*NO emissions:*

*Also the sites with highest deviation of measured and simulated fluxes are among the ones with lowest pH values and highest free Fe. The ForestDNDCtropica model considers explicitly chemo- denitrification based on results revealed from a detailed laboratory study of Kesik et al. (2006). However, lowest pH values in that chemostat experiment was 3 and results obtained were based on cultures of Alcaligenes faecalis. Therefore, it is likely, that the ForestDNDCtropica parameterization of chemo-denitrification is leading to underestimation of measured NO emissions at low pH values. Due to the assumed exponential function this can lead to significant deviation of simulated and measured values.*

Fifth, I would like to see more comparison of the investigators' work with studies by other groups working in upland/montane tropical environments, e.g. Ed Veldkamp, Michael Keller and Whendee Silver's work in other parts of Latin America, Africa and SE Asia. The authors have tended to cite their own work, and the work of Peter Vitousek and Pam Matson, but have not been fully comprehensive in their citations or

inter-comparisons.

*Response: we agree with the reviewer and appreciate his/her suggestion. Now, we have added a comparison with other studies in Latin America. Also, we made more comparisons with forests in China, Australia and Africa (line 376-384).*

Last, the authors need to be more careful in the editing of the manuscript, as there are a large number of typographical and grammatical errors (see SPECIFIC COMMENTS below). Other remarks or observations echo the other referees' comments (e.g. is chemo-denitrification explicitly modelled in this study? Concerns about circularity of the reasoning, etc.)

*Response: we did our best to improve the editing of the manuscript. Chemo-denitrification is explicitly modelled for NO, but not for N<sub>2</sub>O. We do not agree on the circularity of reasoning and refer for a detailed answer to our reply to reviewer 3.*

#### SPECIFIC COMMENTS

1. PAGE 1485, LINES 8-10: "The importance of tropical forests as sink and source of carbon GHGs is relatively well known..." This is overstated; I would suggest deleting or rewriting this sentence. While we have a better understand of the balance of CO<sub>2</sub>, our understanding of CH<sub>4</sub>, CO, volatile organic C (VOCs), black C, organic aerosols and halocarbons is very poor, possibly even worse than our understanding of N<sub>2</sub>O and NO<sub>x</sub>, so this statement is inaccurate.
2. PAGE 1485, LINE 23: Line should read : and denitrification when nitrate is used as..."
3. PAGE 1485, LINE 27-PAGE 1486, LINE 3: This sentence needs to be edited as it is grammatically incorrect.
4. PAGE 1486, LINE 22: Should read "no reliable spatially explicit predictions..."
5. PAGE 1489, LINES 8-11: See comment above. How robust is this 'pedo-transfer function'? What does it actually do? Given how important BD is for the sensitivity of the model, this approach needs to be defended more robustly.
6. PAGE 1490, LINES 11-12: Another typo/grammatical error; this sentence needs to be finished!
7. PAGE 1497, LINES 6-8: Poor grammar/sentence construction. Are these data supported by field studies from the Nyungwe region, or from other tropical field sites? This is left ambiguous.
8. PAGE 1501, LINES 11-12: Grammatical error; this sentence should read "Although there is still considerable uncertainty associated with our emissions estimates, our results provide the first spatially explicitly predictions..."

*Response: the specific comments are all definitely very appropriate and were thus addressed in the revised version.*

#### **Reviewer#2 J. van Haren**

In this manuscript Ghehi et al. combine a process model (ForestDNDC-tropica) with a spatially intensive soil property database (part from the literature, part self generated) to estimate the magnitude and spatial variability of soil NO and N<sub>2</sub>O fluxes in a forested region of Rwanda. They then test whether several soil property data collections over time have much influence on the predicted fluxes.

This paper is interesting and has value based on the regional soil property database

and its value to improve model predictions by its more detailed scale than global soil datasets. However, several aspects of the paper greatly distract and diminish the value of the paper:

1) The model output is not well validated against field data: f.i., the biomass estimates are only roughly compared to one site within the study region,

*Response: this validation criticism is in part already answered in a reply to reviewer 1. With respect to biomass estimates we provided information about the LPJ-Guess model, which is used to initialize the standing biomass and the litter carbon budget in section 2.3.3 (vegetation data). For our study area very limited biomass data is available and we therefore used state of the art biomass modelling to derive initial vegetation and litter conditions for the DNDC model.*

*We ran the model LPJ-Guess in DGVM-mode with default global PFT settings (approx. 10 global PFTs). For the study site, the tropical evergreen PFT was dominant. We did not parameterize a dedicated tropical mountain PFT, but, due to regional climate variations admixtures of temperate tree species were simulated by LPJ for higher altitudes, reflecting the occurrence of limiting temperature conditions during establishment of tropical trees. This leads to a higher fractional cover of temperate trees at higher altitudes and with lower temperatures.*

*Based on personal experience with ForestDNDC(-tropica) forest modelling by various of the authors (Kiese, Werner, Butterbach-Bahl), initial biomass has only a small effect on trace gas emissions simulated by ForestDNDC-tropica as major plant vs. soil microbe competition for nitrogen is mainly occurring at early successional stages but with well-established forests this ratio of N access will remain relatively constant over time. Furthermore, a poor initial biomass estimate will have only small impacts on trace gas emissions simulated by ForestDNDC-tropica for relative short simulation periods, which was proofed by the sensitivity study where wood mass and leaf mass revealed to be of minor and medium importance.*

*We did not validate the LPJ biomass output, as we deemed it unrealistic given the diverse area covered in this exercise. For certain sites the aboveground biomass estimates were very high (up to 400 tons C/ha). For a limited number of locations we could check modelled biomass against measured biomass data from Nsabimana (2009) (PhD thesis), and were found to be of the same order of magnitude as reported from the field. Modelled biomass data for the Nyungwe were also comparable with those reported by Werner et al. (2007) for a forest in Kenya.*

2) the model gas flux output is only validated against soil gas flux incubations, not actual fluxes in the field,

*Response: Please see our first reply to reviewer 1.*

3) Although the authors have detailed soil data, but no detailed climate data is available or incorporated, which I presume can be taken from the global TRMM database.

*Response: We agree that the objectives of the manuscript were challenging and that the subject involved substantial limitations (scarce data, model validation, etc.), but we believe that we did a strong effort to achieve the best possible results by combining one of the best available datasets (for tropic forests worldwide) and modelling tools. Although current TRMM rainfall data at its highest spatial resolution is published at 0.25x0.25 degrees, it was still considered inferior to station data for this study. As the simulation domain is relatively*

*small(0.025x0.025) and the model requires temperature data which is not provided in the TRMM archive, thus requiring data fusion to combine all required weather data for the model, we decided to use weather data from three climate stations, which are at <5km distance from the Nyungwe forest. In addition we used climate data from one recently installed climate station in the Nyugwe forest.*

4) model parameter sensitivity analysis was already done by Kiese et al. 2005 and Werner et al. 2007, I do not see the need to keep repeating this for each model study, unless the model has substantially changed.

*Response: We agree with the reviewer. But, to the best of our knowledge, the influence of individual parameters of Forest DNDC-tropica on the NO flux output on a regional scale has not been investigated before. So, we preferred to do a sensitivity analysis for both NO and N<sub>2</sub>O.*

5) the supplement supporting the hypothesis that high soil NO and N<sub>2</sub>O fluxes could be due to chemodenitrification is very poorly written and the analysis associated with the incubations poorly conceived.

*Response: We improved the information given in the supplement. We acknowledge at the same time that this information is still fragmentary and are currently developing some detailed experiments to further underpin the role of chemo-denitrification and Fe-Anammox for N<sub>2</sub>O and NO emission. We improved the writing style and editing of this part.*

6) The reported gas concentrations in the supplement appear erroneous (5-30 ppm reported for CO<sub>2</sub>, whereas atmospheric values are ~390ppm; 100-200 ppb reported for N<sub>2</sub>O atm ~320ppb), unless the incubation air was treated without mentioning in the text

*Response: We corrected the scale of the figure S1 (CO<sub>2</sub> concentration is ppm). Indeed, all incubation N<sub>2</sub>O and NO data were subtracted by background N<sub>2</sub>O and NO concentrations (so we report the net concentration increase).*

7) The evidence presented in the supplement is suggestive for NO, but not convincing of the production pathway, that the experiment set out to accomplish, by the use of <sup>15</sup>N NO<sub>2</sub><sup>-</sup>, but <sup>15</sup>N NO was not measured (this could have been accomplished by better planning)! This suggests that you did not have NO derived from NO<sub>2</sub><sup>-</sup>, or at least not from the labeled NO<sub>2</sub><sup>-</sup>, which would be the real test whether they were derived from chemodenitrification.

The concentrations reported can be measured in stable isotope laboratories, so I wonder why you say they were below detection limit. Sounds like this part of the experiment was not well thought out.

*Response: We follow the reviewers comment in part: 1) measuring <sup>15</sup>N in NO is not so straightforward, both analytically and in terms of the amount (not concentration) of NO needed). This experiment was carried out at the stable isotope lab of the last author.*

*Moreover we observed that the NO and N<sub>2</sub>O production occurred very fast (which is also an indication for abiotic production) and when we extracted the soil and the end of the incubation we did not detect any <sup>15</sup>N in NO<sub>2</sub><sup>-</sup> anymore, which indicates that all NO<sub>2</sub><sup>-</sup> added has been removed. We want to point again to the reviewer's attention that these incubations were carried out in sterilized soils, so biotic N-gas production is excluded.*

This paper would be greatly enhanced if the authors could include chemodenitrification in their model to test whether that indeed can resolve the poor flux predictions by the current model version. However, the poor quality of the presented data in the supplement leaves me skeptical whether this is useful and the quality of the dataset the model is being tested against.

*Response: In the revised version, it is mentioned that chemo-denitrification (only effect of pH) has been included in the ForestDNDC-tropica model only for NO (mainly based on a lab experiment of Kesik et al., 2006), but not for N<sub>2</sub>O. However, the design of our current experiment and data does not allow us to further refine the algorithms of chemo-denitrification in the model, i.e. we need more data for calibrating the model in particular with low pH values and high Fe like in our experiment, however, this was not the aim of this study.*

*In this part of our study we only put forward some hypothesis that could be the subject for future research. Thus, the evidence that we present here is preliminary, but sufficient (seven soils were used and we provide evidence for NO and N<sub>2</sub>O formation via an abiotic pathway) to formulate a plausible hypothesis that is worth for future testing, i.e. the role of chemo-denitrification and FeAnammow (see Yang et al 2012) as these acid and Fe-rich soils at least have the required soil conditions for both processes*

Please also note the supplement to this comment:

<http://www.biogeosciences-discuss.net/10/C208/2013/bgd-10-C208-2013-supplement.pdf>

Figure S1 No explanation in the text how you got the replicates for the error bars

*Response: in the supplement, we noted that all seven soils samples were incubated in three replicates. So, the error bars are plus one standard deviation of these three replicates*

The data represented in graph S2 appear to contradict the fluxes reported in table S2. Please explain.

*Response: we expressed and calculated the N<sub>2</sub>O and NO incubation fluxes in  $\mu\text{g m}^{-2} \text{hr}^{-1}$  in order to compare these data with other soil incubation fluxes (e.g. Gharahi Ghehi et al., 2012) or field fluxes.*

*In this calculation the ideal gas law in combination with the molecular weight of N<sub>2</sub>O and NO was used to calculate the fluxes in  $\mu\text{g N}_2\text{O l}^{-1}$  and  $\mu\text{g NO l}^{-1}$ . Those values were then recalculated into  $\mu\text{g N}_2\text{O h}^{-1} \text{kg}^{-1}$  dry soil and  $\mu\text{g NO h}^{-1} \text{kg}^{-1}$  dry soil by using changes in headspace concentrations over time using a linear regression approach. The NO and N<sub>2</sub>O  $\text{h}^{-1} \text{kg}^{-1}$  dry soil fluxes were finally recalculated to  $\mu\text{g N}_2\text{O m}^{-2} \text{h}^{-1}$  and  $\mu\text{g NO m}^{-2} \text{h}^{-1}$  applying a known soil surface area.*

*It has been mentioned in the manuscript that we applied the same laboratory techniques that were used by Gharahi Ghehi et al. (2012a) to measure soil N<sub>2</sub>O and NO production. The soil samples were incubated in the laboratory in tubes of 2.6 cm diameter, 9 cm height and 47.78cm<sup>3</sup> volume.*

*We are thankful for the short comments. All were corrected in the revised version as the reviewer suggested or answered (the above referee's reply).*

### Anonymous Referee #3

1) Sections 2.6 and 3.3 are identical. Why?

*Response: We corrected section 3.3 in the revised version.*

2) In several places, the argumentation is circular. One example is at the end of page 1498, beginning of page 1499: “For instance, N<sub>2</sub>O emissions exceeding 4 kg ha<sup>-1</sup> yr<sup>-1</sup> were found in the northwestern part of the forest, which is characterized by high clay and OC contents and low pH (<4). Several authors (: : :) have shown that low pH decreases the activity of the N<sub>2</sub>O-reductase, thereby increasing production of N<sub>2</sub>O, rather than N<sub>2</sub> from denitrification. For nitrification, it has also been demonstrated that low pH<5 or=4 values favor N<sub>2</sub>O production (: : :)”

Circularity of the argument lies in the findings of the cited authors (or similar studies) having been incorporated into the model, which in return confirms these findings. Consequently, these model results are no new information. They merely confirm the model operates as it has been asked to operate.

*Response: We had 3 objectives for our study: 1) predict the N<sub>2</sub>O and NO source strength of a tropical mountain forest soil using a process based model tested with results from an incubation experiment; 2) to what extent do these values differ from current estimates; and 3) how sensitive is ForestDNDC-tropica model to changes in driving input variables.*

*We do not agree that our argumentation is built on circular argumentation! The ForestDNDC-tropica model as any model (either empirical or process based) does NOT operate how it is “asked to operate” but is developed, refined and parameterized by using results from process studies and field experiments on C and N turnover and fluxes and their environmental controls for tropical forest ecosystems. Hence we argue that our argumentation is rather based on the “logic of building a model” and not on circularity. However, data availability of tropical forest on N trace gas emissions is still very scarce and therefore current state of the model and parameterization has some degree of uncertainty. For that reason it is not obvious that a model developed based on data of other sites is performing well at a new site. In this respect our finding that the current version of the model may underestimate NO emissions via denitrification is very important and triggers future experiments, which indeed can then be used to further refine mechanistic descriptions and parameterisation of important processes involved in N trace gas production, consumption and emission.*

3) Conclusions: page 1501, last sentence: “In particular, chemo-denitrification processes on acidic soils seem to be under represented in the current ForestDNDCtropica model.” This conclusion is solely based on one of the authors’ previous studies (see page 1499, lines 15-17: “Furthermore, Gharahi Ghehi et al. (2012a) suggest that high N<sub>2</sub>O and NO emissions for some sites in the Nyungwe forest are possibly due to chemodenitrification processes.” Why does it feature as a conclusion drawn from the work presented in the current manuscript? What has the current study contributed to support this conclusion?

*Response: Indeed we concluded that the ForestDNDC-tropica model at present state may underestimate N<sub>2</sub>O and NO fluxes since preliminary data suggest that abiotic processes responsible for NO (especially) and N<sub>2</sub>O formation might be important for the soil types of Nyungwe (high in Fe, very acid pH) and are not well enough parameterized in the model. We provided evidence of abiotic NO and N<sub>2</sub>O formation in the supplementary information.*



*In addition to the availability of detailed information on soil, vegetation and climate data (which in itself is challenging for the tropics especially in Africa) and that was the main reason why we have chosen the Nyungwe forest for which, compared to other locations, a good soil and climate data base is available). We believe that ForestDNDC-tropica model which originally was adapted from the temperate ForestDNDC model requires future improvement to encompass all biotic (e.g. dissimilatory nitrate reduction to ammonium) and abiotic (e.g. chemo-denitrification, Feammox) sources of N<sub>2</sub>O and NO emissions which are particularly important in wet tropical systems.*

I am sure the senior authors of this study will find more examples of circular arguments and inappropriate statements, once they carefully read their manuscript. Equally, I am confident that they are able to make appropriate changes and corrections.

*We assure the manuscript has (and was) carefully checked (also by the senior authors). To the best of our knowledge we have modified the manuscript according to all suggestions made by the referees and improved/removed circular arguments. We removed inappropriate statements, but (as mentioned above) are not convinced that with respect to the development, further refinement and application of process based models such as LandscapeDNDC our argumentation is circular.*

## References

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*oxidation coupled to iron reduction. Nature Geoscience, 5, 538–541, doi:10.1038/NGEO1530, 2012.*