Author's Response

Responses to B. Stocker (Referee)

We would like to thank B. Stocker very much for his helpful suggestions and interests in our manuscript. We list our opinions point-by-point in response to his comments or suggests. Modifications to the manuscript can be tracked in the submitted MS.

SUMMARY

This paper describes the implementation of a model for inorganic soil nitrogen (N) dynamics within a Global Dynamic Vegetation Model that explicitly treats the interactions of the carbon (C) and N cycles. Results are presented from a simulation covering years 1970-2005 and for several sensitivity analyses (soil moisture, elevated CO2, warming). The model is assessed against observational data of N2O emissions from a set of observations that are collected for the present study. Apart from confirming global total N2O emissions are on the same order as previous studies suggested (the central estimate here is 6.82 TgN2O-N/yr), the authors conclude that "Improvement of soil hydrology is likely to significantly reduce the large uncertainties associated with soil N2O emission estimates".

Response: Thank you for taking time reviewing our paper.

This is a straightforward and honest model description and presentation of its performance and presents some valuable insights into the general model behaviour in response to basic environmental drivers (CO2, warming, combination of the two). This is essential for the interpretations of model results also in view of future studies addressing N2O emissions conducted with this version of the LM3V-N model. Benchmarking model performance and a concise description of implemented code should be considered best practice and the study presented here is a good attempt at this ideal.

Response: We appreciate the reviewer's positive comments on our study.

But does it convincingly succeed at thoroughly describing the parameterizations and benchmarking the model performance? In this respect, I have some concerns which should be addressed in a revised manuscript. The present study may warrant publication if the authors address the issues raised below.

Response: We have carefully considered all issues raised by this reviewer. Responses and revisions are provided accordingly.

In summary concerns are: - Concerning difficulties of benchmarking a coupled system:

Did the authors really look at the most important factors determining N2O emissions?

- The authors did not attempt to decouple their new implementation of inorganic N dynamics from the behaviour of other model parts in which their "module" implemented.

Therefore, results are subject to these other model parts.

Response: We agree that N_2O emission from our study is subjected to the performance of other model parts. Particularly the model is sensitive to processes that allow buildup of inorganic or mineral nitrogen (ammonium and nitrate), which happens if nitrogen (N) is limiting for decomposition and plant growth. The sensitivity to N limitation is due to the fact that denitrification is considered as a "weak" sink, where removal coefficients of plants and soils are much higher if there is sufficient demand for N. The second sensitivity is the fraction of N_2O generated during nitrification. In this vein, we add sensitivity tests in sect. 2.2.3 and sect. 3.4 We now investigate model performance under altered N input from fixation, and changes in other N fluxes that affect inorganic N dynamics, the concentration of inorganic N in soils, and thus denitrification. These changes are associated with the following hypotheses: 1) The change in N fixation from a dynamical model that responds to N limitation to a static model based on reconstruction has the potential to add N critically above what is needed and remove a negative feedback that is inherent to LM3V-N. This addition also moves the dynamics of LM3VN towards schemes used in other models, where N fixation is scaled to net primary productivity (such as CLM), or transpiration (such as ISAM). 2) The sensitivity of excluding dissolved organic nitrogen reroutes some of the N that would be lost as organic form (DON, fire) through the mineral pool, and can therefore increase N2O emissions 3) Reduction in plant uptake strength leaves more available N for leaching and denitrification, or in other words increases the relative sink strength of denitrification vs. plant removal. 4) The parameter that determines the gaseous loss during nitrification can ultimately shift the competition for the overall available N because it removes N before it becomes available as nitrate. 5) allowing all N from fire to remain in the plant soil system also reduces unavailable losses, and increases potential denitrification losses due to resynchronization of plant demand and mineralization,

and the overall fact that more N is retained in the system. Overall, these sensitivity experiments test how denitrification plays out within the larger soil-plant system and how the larger N cycle is linked to denitrification.

- Presentation: For a model description and benchmarking exercise like the present study, the journal Geoscientific Model Development would suit even better than Biogeosciences. - The authors implemented a "module" for inorganic N dynamics, but the paper focuses only on N2O emissions. However, N2O emissions are governed by the inorganic N dynamics. Regarding the aim of this paper (model description/benchmark) these other processes warrant equal weight.

Response: We appreciate the reviewer's suggestions for Geoscientific Model Development as a better choice. We have carefully considered the reviewer's suggestions, but we would like to emphasize that model development and benchmarking is not the only focus of our study. In particular, the denitrification scheme we implemented is used in other models. We think the question we are asking are more of what is the result of established denitrification routines into a different model knowing that each model has a different "philosophy" of the larger plant-soil N cycle. We also think that the comparison against different site should not be the central part, but we would like to put emphasis on the question what happens if one implements established denitrification routines into a different model. How does it impact N_2O budget, N_2O fluxes at different sites, the response to global change factors, and to how the water cycle is treated? Emphasizing that our model is not new, we think our attempt to answer these questions fits into Biogeoscience. In that sense we appreciate Beni's suggestion to evaluate N_2O fluxes with the larger N cycle in mind.

GENERAL COMMENTS

WHY BIOGEOSCIENCES?

The present study would fit the scope of Geoscientific Model Development (another open-access Copernicus journal with a high impact factor) perfectly. This would allow for a better reproduceability, re-usability and tractability of code developed here. GMD requires model code to be made public. Of course, making the entire LM3V-N code public may not be practical here and I am aware of the challenges of de-coupling individual model parts that are usually

run in tight coupling with other model parts. However, this should not prevent development of parts of larger models to be published in GMD. A practical solution may be found to provide developed code as a module and some overhead to drive that module in a "demonstration mode". Could that be achieved? In this case, I strongly recommend publication in GMD. This is the best way to share innovations, advance science (and even get more citations). Also the data in Table B1 could be made publicly available in a convenient format. GMD provides a great platform to share such data.

Response: We share the reviewer's opinion that public code ultimately is a great tool to advance science. We also have the reservation he mentioned, with respect to the practicality of doing this work. LM3V-N has a significant overhead and its implementation on a new platform is complex. The solution of a demonstration code could be an alternative, but since the nitrification-denitrification code is not new per se (just implemented in a different model) we feel there is little to be gained from a demonstration module. We appreciate the reviewer's suggestions for Geoscientific Model Development as a better choice. We have carefully considered the reviewer's suggestions, but think Biogeoscience fits our study also.

CHALLENGES OF BENCHMARKING A COUPLED SYSTEM

Paper deals with a process (N2O emissions) that is very challenging to model. This is because of the C-N cycle system dynamics with "circular coupling" where response time scales of individual processes determine the system response on different time scales. It is inherently difficult to thoroughly benchmark such a coupled system. The challenge is that N2O emissions are dependent on all aspects of the C-N cycle.

Response: We appreciate the reviewer's acknowledgement of the challenges associated with this study, we have amended the text with insights how LM3V operates with respect to other parts of the N cycle in the method section. We expanded sensitivity tests that include now fixation, fire, DON losses, and plant uptake, next to the "classical" nitrification-denitrification parameters.

The study presented here appears to be subject to these problems as well. Benchmarking individual processes in a coupled system without actually de-coupling separate model parts

may be misleading. In some instances (e.g., correlation analysis, Sect. 3.4; strong focus on sensitivity to WFPS) the analysis presented here is subject to this problem and it is confusing in what insight some analyses really provide.

Response: We agree that correlation analysis does not provide much information and is removed from the revised version of the manuscript. However, we think WFPS is an important factor contributing to the uncertainty of terrestrial N_2O simulations and is one of the focuses of this study. Because nitrification-denitrification requires spatially or temporal conditions alternating aerobic and anaerobic conditions, which is parameterized via WFPS, this is probably the single most important factor, after the N requirements of plant and soil have been taken care of. With the improvement of the manuscript, by considering other factors, we can put the sensitivity to WFPS in a much better context.

In my understanding, N2O emissions are determined by two (largely independent) aspects:- denitrification/nitrification throughput; This scales linearly with substrate (nitrate and ammonium) pool size (their Eq. A1 and A4) which in turn this is governed by the balance of net mineralisation, plant N uptake and losses. It is thus affected by the whole system of C-N interactions. Benchmarking this aspect of N2O emissions thus requires a wide focus of benchmarked quantities. - fraction of N2O lost with denitrification/nitrification. This is determined by soil oxidation availability (their Eq. A8 - A11).

This fraction is relatively uncertain.

Response: We agree that N_2O emission is affected by the whole system of C-N interactions, and is one of the reasons or advantage for us to analyze N_2O emissions within a global C-N model. The fraction of N_2O lost during nitrification is set as a constant of 0.4%. We test the sensitivity of N_2O emissions to this fraction by setting this value to 4% or 0.04%. The model is most sensitive to this fraction compared to parameters regulating plant N uptake, nitrification and denitrifiction rates. However, this fraction is very uncertain based on limited field or laboratory studies. Goodroad and Keeney (1984) suggested a value of 0.1-0.2% , while Khalil et al. (2004) reported a range of 0.16%-1.48% depending on the O_2 concentration. We applied a value of contant 0.4% in the default run which might cause large uncertainties in our results. The fraction of N2O lost from denitrification is taken from the empirical estimation from the DayCent model (the daily version of the CENTURY model), and has been assesed under

different conditions (Del Grosso et al., 2000). DayCent has been widely applied in trace gas studies across terrestrial ecosystems. This fraction also embraces large uncertanties. We acknowledge this fact in our improved manuscript.

Thus, the challenge is that N2O emissions are dependent on all aspects of the C-N cycle. Soil moisture affects the amount of inorganic N subject to denitrification and nitrification. The strong focus of this study on assessing the model sensitivity to soil moisture (_water-filled pore space, WFPS) is thus questionable.

Response: We agree with the reviewer that N_2O emissions are dependent on various aspects of implementation of C-N cycle in our model. Based on the reviewer's suggestion, we add a comprehensive set of sensitivity tests in sect. 2.2.3 and sect. 3.4 (***Sensitivity to N cycling processes and parameterization***) to analyze the influence of N cycling processes other than nitrification and denitrification on N_2O emissions. These tests include effects of biological N fixation, DON losses, fire, and plant uptake capacity.

The authors implemented a full representation of inorganic soil N dynamics (p.3106, l.1: "Here, we add a soil nitrification–denitrification module"). However, this paper puts a very strong focus on N2O emissions. As mentioned above, N2O emissions are governed by the inorganic N dynamics. I think, benchmarking N2O emissions would be more powerful, if observational constraints on other quantities determining the inorganic N dynamics of different levels be included. Examples of such quantities are: -inorganic N pool size (given net mineralisation rates) - N loss rates (given inorganic N pool sizes) - nitrification/denitrification rates (given inorganic N pool sizes) – sensitivity of nitrification/denitrification rates to different soil conditions.

Response: Agree. The focus of this manuscript is on N_2O emissions. However, N_2O fluxes are strongly regulated by inorganic N dynamics. It is beneficial if other quantities regulating inorganic N dynamics are validated. One of the biggest factor that sets inorganic pool sizes are the sink strength of plant uptake (under N limitation), and the presence/absence of N limitation (or in other models, how plant N status affect uptake). Soil N pool measurements can be helpful as an additional benchmark. However, as the author noted, it is the larger plant soil N cycle, including that sets plant N demand, and N limitation. These questions are great challenges for

all models. Unfortunately, we do not have large scale observation data available for benchmarking the global model with regard to quantities such as inorganic N pool size, nitrification and denitrification rates.

SUBJECT TO PERFORMANCE OF LM3V-N

This is in some respect related to the comments raised above. The authors test the model part representing inorganic N dynamics, as implemented in the LM3V-N model. However, some sensitivity analyses presented here are tightly dependent on the sensitivity of the LM3V-N model (Sect. 3.5). This requires at least a description of the general functioning of that model (How are major N input and loss fluxes represented? What leads to N limitation? What governs N fixation?)

Response: Agree. Further description of the general N cycle is added to sect. ***2.1.1 Main characteristic of LM3V-N***. And we discuss it with respect to our newly added sensitivity analysis. Adding an established nitrification-denitrification model is indeed subject to the overall "philosophy" of the entire biogeochemisitry model. We believe, it is thus useful to evaluate N2O emission in this sense.

In my understanding, with inorganic N dynamics represented broadly equally (which is the case for all global vegetation models that simulate C-N dynamics and N2O emissions: DyN-LPJ, Xu-Ri et al., 2012; LPX-Bern, Stocker et al., 2013; O-CN, Zaehle et al., 2011), N2O emission sensitivity to CO2 and warming primarily depends on the degree of progressive N limitation under environmental change (less N2O emitted in a N-scarce system). Here, these models' predictions diverge substantially. On one side,O-CN generally more N limitation under elevated CO2 (=increased plant demand), on the other side DyN-LPJ and LPX-Bern (pretty much the same) does hardly generate N limitation on a decadal time scale. This model behaviour is contingent on how N inputs into the system are simulated (we know that losses are broadly equal as they all rely on a DNDC-type model for inorganic N dynamics). O-CN simulates BNF using an empirical relationship with evapotranspiration. DyN-LPJ implies a BNF flux by holding soil C:N ratio constant, i.e., higher litter-to-soil C flux implies additional N brought into SOM, which is ultimately made available for plant N uptake after mineralisation. To interpret the results presented here, it is crucial to understand where in this spectrum of O-

CN and DyN-LPJ this model is. The information provided in Sect. 2.1 ("BNF in LM3V-N is dynamically simulated on the basis of plant N availability, N demand and light condition.") doesn't provide sufficient insight to understand this crucial model characteristic.

Response: The reviewer is spot on. BNF in LM3V-N is different from that of O-CN, LPJ-DyN and LPX-Bern. Further description related to BNF is added to sect. 2.1.1.5. And more details are available in Gerber et al. (2010). BNF in LM3V-N is active only when plant N requirement is failed to be satisfied by root uptake, and is adjusted according to plant N demand. LM3V-N assumes a tighter (smaller input and smaller losses) in preindustrial N cycling compared to O-CN, LPJ-DyN and LPX-Bern with smaller amount of BNF (72 in LM3V-N vs. 104 TgN yr^{-1} in O-CN) (Zaehle et al., 2010). The adaptive BNF also contributes to the tighter N cycling. However, in conditions of N limiation, there is considerable adjustment of BNF in response to progressive N limitation, as illustrated by ca. 2 times increase averaged over 100 years (Panel (3), Fig.8) under doubling of atmospheric CO_2 level. This strong negative feedback via BNF alleviated N limitiation intially faced by tropical forests, and turns the negative N2O response to positive after several decades. Deference in BNF is one of the major causes of divergent responses to CO2 fertilization between those models. Results related to BNF is added to sect.3.4 , 3.5 and discussion of the revised manuscript.

CORRELATION ANALYSIS IN SECT. 3.4

Are correlations derived from regressing the corresponding time series of the historical run? Temporal resolution (daily/monthly/annual)? I'm a bit confused about what such a correlation actually represents. Short term correlations don't necessarily represent the system's sensitivity to a certain input. I guess that's really what you are after here: understand the characteristics of the model - its sensitivity to different driving variables. Isn't this better covered by your analysis of step changes? The analysis presented here is particularly confusing in the case of the correlation between N2O emissions and Ammonium. I'm pretty sure that, if you would add a certain amount of Ammonium everywhere (N fertilization experiments), N2O emissions would increase not decrease - also in the model presented here. The temporal correlation presented here thus does not provide direct insights into the model sensitivities. I think, the confounding aspect is that there is also a time-scale dependence of such correlations (delayed response of some variables in the system). Another aspect that is confusing about the analysis presented in

Fig. 5 is that some correlations are with variables that are directly or indirectly external to C-N cycling (temperature, soil moisture, GPP), while others are intrinsic quantities (nitrate, ammonium, etc). Regarding the negative correlation of N2O emissions with ammonium concentrations: This is confusing as Eq. A1 says that nitrification (_N2O emissions) and ammonium are directly proportional. I suspect that this counter-intuitive result is due to the fact that ammonium levels are low in the tropics due to the high plant N demand. At the same time, also net mineralisation rates must be quite large (is that so?) and nitrification rates must be high as well which implies high N2O emissions. Is the result presented here really indicative of what's driving N2O emissions?

Response: We Agree. The correlation analysis may provide some insights, but confuse when discussing mechanisms. The response to ammonium availability is such an example. On a side note, because nitrification is strong in LM3V compared to other sinks, any increase in nitrification strength will draw down ammonium concentration.

MODEL DESCRIPTION IN APPENDIX

Appendix A contains "the heart" of this paper. This paper is primarily a model description and benchmarking exercise. The model is not applied to address a specific question or a particular period. I find it inconsistent with the scope of the paper, to put the actual model description (the "heart") into the appendix.

Response: We gladly follow the reviewer's suggestion and move the appendix into the method section in the main text (sect ***2.1.2 Soil N_2O emission***).

"Our simulation of N2O losses during nitrification–denitrification generally follows the "hole-in-pipe" concept". To my understanding, this concept refers to models that assume that gaseous N losses are proportional to net mineralisation rates. The model presented here assumes that N losses are scale with inorganic pool sizes (proportionally for nitrification - not really a loss term though) and with Michaelis-Menten kinematics for denitrification (not mineralisation rates). In my understanding, the model presented here is thus not a hole-in-the-pipe model.

Response: The original "hole-in-pipe" model assumes that gaseous N losses are proportional to net mineralization rates. It describes the rate of nitrogen cycle as the amount of nitrogen flowing through the pipes. N_2O leaks out of the pipes depending on nitrogen cycling rate as well as the size of the holes, determined largely by soil water content(Firestone and Davidson, 1989). Our understanding is that this metaphor of "nitrogen flow through the pipe" is not

constraint to net mineralization. Instead, it is generalized to nitrogen availability and can be indicated by various indices such as N mineralization, nitrification potential and the inorganic pool sizes (e.g. Davidson et al. (2000)). However, we agree with the reviewer that our expression is confusing. And "Our simulation of N_2O losses during nitrification-denitrification generally follows the hole-in-pipe concept (Firestone and Davidson, 1989) with more detailed treatment of the N flux pipes and the leaky holes (gaseous losses) in the pipes" is deleted from the revised manuscript.

As a further remark on the "hole-in-the-pipe": Can't we say that the "hole-in-the-pipe" concept is simply wrong? In such a model, N losses are not affected by N demand. That is, if net-mineralisation is increased, losses are increased irrespective of whether demand for N uptake is increased. Hence, warming may not stimulate plant growth (in contradiction with observations) and elevated CO2 will tend to lead to a state of progressive N limitation as N losses are not reduced. Both are not match observational findings (Melillo et al., 2011; FACE results). Further, Davidson et al. (2007) present evidence that N2O emissions are indeed reduced when demand outweighs net mineralization and leads to depleted inorganic N pools. Maybe add this to discussion.

Response: We agree. The mineralization based approach is added to Discussion, which states "If gross mineralization is used as an indicator of the rate of N flow in the "hole-in-the-pipe" concept and gaseous losses are propotional to mineralization, the inital negative response is unlikely to be detected. We found increased mineralization rate with increased litterfall under elevated CO_2, while N availability is reduced from LM3V-N. The mineralization based approach is likely to predict an inrease of losses regardless of N limitation".

SPECIFIC COMMENTS——————————p.3102 l.3-5: "With high temporal and spatial heterogeneity, a quantitative understanding of terrestrial N2O emission, its variabilities and reponses to climate change is challenging." â˘A˘Tre wording to "Due to its high temporal and spatial ..."

Response: Agree. Change is made to P1 line 10 of the revised manuscript.

l.9: state explicitly if you applied the model to sites pecific driving data or extracted the corresponding gridcell's output

Response: Correct. We extracted the corresponding gridcell's output. Explicit explanation is added to p.1 lines 16-17 of the revised manuscript, which says "Results extracted from the corresponding gridcell (without site-specific forcing data) was comparable with the average of cross-site observed annual mean emissions"

l.11-15: State the response of N2O to elevated CO2.

Response: Corrected. The revised manuscript states: We found that the global response of N2O emission to CO2 fertilization was largely determined by the response of tropical emissions with reduced N2O fluxes in the first few decades and increases afterwards. The initial reduction was linked to N limitation under higher CO_2 level, and was alleviated through feedbacks such as biological N fixation. The extratropical response was weaker and generally positive, highlighting the need to expand field studies in tropical ecosystems.

p.3103 l.1: You may state the contribution of N2O to total anthropogenic radiative forcing.

Response: Agree. Add to p.2 lines 14-15 of the revised manuscript.

l.4: Unclear what you mean with "comparable to the combined anthropogenic emissions"

Response: Agree. We deleted "comparable to the combined anthropogenic emissions" to reduce confusion. See p.2 line 17 of the revised manuscript.

l.20: 'particularly' instead of 'particular'

Response: . Change made to p.2 line 31 of the revised manuscript.

p.3104 l.18: In my reading, LPJ DyN simulates a positive response of global N2O emissions to CO2 (blue line is above purple line in Xu-Ri et al. (2012), Figure 5).

Response: CO_2 plus interaction with climate result in a positive response of global N_2O emissions in Xu-Ri et al., (2012), but historical CO_2 change alone (single factor, from Fig. 7 of Xu-Ri et al., (2012)) causes a slight decrease in historical N_2O emissions. To clarify, we rewrote this part as: Simulations with O-CN demonstrated a positive response of N_2O emissions to historical warming and a negative response to historical CO_2 increase, globally. While CO_2 and interaction with climate change resulted in an increase in historical and future N_2O emissions from LPJ-DyN (Xu-Ri et al., 2012) and its application (Stocker et al., 2013), respectively, historical CO_2 change alone (single factor, from Fig. 7 of Xu-Ri et al., (2012)) caused a slight decrease in historical N_2O emissions.

You may also want to refer to Stocker et al., 2013: N2O response from another implementation of Xu-Ri's adaptation of DNDC.

Response: We added the omitted reference to p.3 lines 21-23,27-31 of the revised manuscript.

l.21: Xu et al., 2012 is usually referred to as Xu-Ri et al., 2012 (see references 'Xu-Ri & Prentice, 2008' in her own publication Xu-Ri et al., 2012).

Response: Corrected.

l.29: "data-overriding" Can you explain this differently - wasn't clear to my first reading.

Response: Agree. "Data-overriding" is changed to "replacing the model soil moisture"

p.3105 l.11: Does LM3V-N use fixed prescribed C:N ratios in different compartments?

Response: Yes. LM3V-N uses fixed prescribed C:N ratios in different compartments. In addition, LM3V-N has a N storage pool that buffers asynchronies in C and N dynamics. For more details, please refer to 2.1.1.1 C-N coupling in vegetation or Gerber et al. (2010).

Please clarify. Sect. 2.2.1.: Good, accurate description.

Response: We have trouble finding this expression in this section but stand by to make any further clarification.

p.3108 l.16: do you really mean "maximum"?

Response: No. Maximum is deleted and text is rewording to "LM3V-N uses the concept of plants available water, where the water that is available to plant varies between the wilting point and field capacity".

l.25: I'm confused, units don't add up. Also, it is unclear where other parameter values in Eq. 1 are derived from. Eq. 1 is the only equation presented in the main body of the manuscript, yet it describes a quantity of secondary (if not tertiary) importance (WFPS -> rates -> N2O emissions). This appears somewhat inconsistent with the presentation of more important equations only provided in the Appendix.

Strong emphasis is put on assessing different formulations of WFPS, yet an function of WFPS is actually applied for determining denitrification/nitrification/volatilisation rates and NOx:N2O partitioning in the model, and this function contains parameters which are not described and assessed.

Response: Further explanation of parameter values and units are added to Eq.1 (Eq. 22 in the revised version). The formulation is revised as:

$$WFPS = \frac{\frac{\theta}{\rho h_r}}{1 - \frac{BD}{\mathrm{PD}}}$$

while *WFPS* is the water filled porosity, θ (kg m^{-2}) is the root zone soil water; h_r (m) is the effective rooting depth of vegetation; ρ is the density of water (1000 kg m^{-3}); *PD* is the particle density of soil (2.65 g cm^{-3}); and *BD* is the bulk density of soil (in unit g cm^{-3}) obtained from the Harmonized World Soil Database (HWSD) version 1.1 (Wei et al., 2014). We add more detailed description of the main characteristic of LM3V-N (sect. 2.1.1) and soil N_2O emission (sect. 2.1.2) to the main text. WFPS is involved in nitrification/denirification/volatilisation as well as the partition of N gases as reported by various field and modelling studies. The NOx:N2O partitioning is taken from the empirical relationship derived by Parton et al. (2001)

which is applied in the daily version of the CENTURY model. These constants are empirically derived based on field measurements. $\frac{D}{D_0}$ denotes the relative gas diffusivity in soil (D) compared to that in the air (D_0). $\frac{D}{D_0}$ is calculated based on air filled porosity. The parameter represent the gas diffusion in air (D_0) is not accually used in calculation. To clarify, we replaced the notation $\frac{D}{D_0}$ by D_r in the revised manuscript (Eq. 14-15).

p.3109 l.17: ": : : field scale." References?

Response: Agree. References "Dijkstra et al., 2012;van Groenigen et al., 2011" are added

l.22: At what point in the simulation does the CO2 doubling become effective?

Response: Here, we evaluate the model's response to step changes in form of a doubling of preindustrial CO2 level (284 ppm to 568 ppm) and a 2K increase in atmospheric temperature.

p.3110 title of Sect. 2.3: Could ": : : with environmental variables" be replaced by ": : : with observations"? This would make more sense to me.

Response: Agree.

p.3111 l.3: Did you get this value spot-on from blindly implementing the equations with parameter values described here or was there any tuning involved? Not that this would be problematic, but it should be mentioned here to provide clarity.

Response: We did not aim at this value. As the reviewers mentioned, this value is sensitive to the fraction of net nitrification lost as N_2O. We highlight this now in the discussion section.

Where does uncertainty range stem from? Why is the uncertainty range not displayed in Fig. 1? Or is it just a range of values for different years. Please clarify.

Response: Based on another reviewer's suggestion, we supplied three budget values corresponding to the three soil moisture datasets. The simulated global soil N_2O flux is

6.69±0.32 TgN yr^{-1} (1970-2005 mean and standard deviation among different years) (Fig.1) with LM3V-SM (Method 3), 5.61±0.32 TgN yr^{-1} with NOAH-SM (Method 2) and 7.47±0.30 TgN yr^{-1} with ERA-SM (1982-2005, Method 2). The uncertainty (±) stands for the standard deviation of N_2O emissions from different years for each soil moisture dataset, which we clarified in our manuscript. Annual N_2O values from different soil moisture datasets are also added to Fig.1.

p.3112 l.10: highly variable savannah emissions: when high/low? during wet season? confusing units (seasonˆ-1)

Response: High emissions are during wet seasons and low emissions in dry seasons. Units are changed to month^-1 instead of season^-1 for Fig.2.

p.3115 l.5: Xu-Ri et al., 2012 suggests positive effect.

Response: Please refer to the answer to an earlier question in p.3104 l.18. To reduce confusion, we removed reference to Xu-Ri et al., 2012 in the revised manuscript.

l.13: "net effect depend on : : :" See my general comment "SUBJECT TO PERFORMANCE OF LM3V-N".

Response: Agree

p.3116 l.18: delete "knowledge from"

Response: Corrected.

p.3117 l.10: Wouldn't such environmental gradients (along which primarily temperature and precipitation change) offer a great testbed for N2O model benchmarking?

Response: Agree. Environmental gradients provide us great opportunity to test the models. Although altitudinal changes result in temperature or precipitation gradients, these gradients are within one model grid cell and the model does not incorporate topographical information explicitly. It is difficult to make use of the altitudinal data.

p.3120 l.9: typo: "speicies"

Response: Corrected.

Appendix in general: Parameter values are presented in Equations without any further description and reference. Can this be improved?

Response: Appendix A is rewritten and moved to sect. ***2.1.2 Soil N_2O emission*** in the main text. We replaced the notation $\frac{D}{D_0}$ by D_r in the revised manuscript (Eq. 14-15). We also add corresponding refereces, units and further descriptions for parameters such as b_{N,NH_4^+}, $b_{NO_3^-}$, k.

Fig.5: I recommend to use a two-colour scale (e.g., blue-red)

Response: According to reviewers' suggestions, we agree that Fig. 5 does not provide much information and delete Fig.5

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Responses to I. C. Prentice

This MS presents an extension to an existing model, allowing the simulation of N2O emissions, which are benchmarked against a newly compiled data set of observed emissions.

Response: Thank you for taking time for reading and commenting on our discussion paper.

I suggest that this work is not yet ready for publication. More work to evaluate and improve the model is required before final publication. When it is finally published, more information should be provided about how the modelled N cycle works, as the basic principles are not clear from this description.

Response: In response to this comment and comments from other reviewers, we moved and amended the appendix with the model description into the main text. We further added additional description of the main characteristic of LM3V-N that we thought may be relevant of the fast processes that govern the dynamics of ammonium and nitrate in soil.

Generally we might expect a publication describing a model to represent an advance in knowledge over the current state of the art. It is not clear to me how this manuscript does so. As one referee (Beni Stocker) has pointed out already, one would reasonably expect to see independent evaluation of various quantities that underlie the process of N2O emission, but this is not provided.

Response: While our work may not be vertical advancement, we add important pieces of evidence that help the scientific community understand how N_2O emissions are dealt with in global models. We clearly state that we build on earlier work that put forward formulation on nitrification and denitrification. It is important that the model setup for the larger N cycle differ from model to model, and thus the implementation of similar mechanism in a different model provide critical insight. In response to another reviewer's comment, we provide a sensitivity test to critical parameters of the larger N cycle, including mechanisms that govern N input and N losses (biological N fixation, DON losses, plant uptake capacity, fire), and discuss these.

The new data compilation, oddly and without explanation, contains only about a quarter of the N2O emissions data previously compiled by Xu-Ri et al. (2012).

Response: For the observational data we compiled, we try to limit the impact of human disturbance such as land use change since we do not conduct land use change simulations in this study. We explain this now better in the revised paper. The larger N cycle responds critically and long-lasting to disturbance (e.g. Bernal et al., 2012). Most of our data are from pristine ecosystem without documented land use change, or at least have no disturbance within the latest 50 years for forests and 10 years for grasslands. Therefore our selection criteria differs compared to Xu-Ri et al. (2012). Despite our careful selection our compilation has 61 data points only 5 less than Xu-Ri (2012).

And when the data-model comparison is made (in Figure 3), the goodness of fit appears to be inferior to that achieved by the model of Xu-Ri et al. (2012). Xu-Ri et al. (2012) also performed a series of sensitivity experiments that showed consistency with a wide range of published experimental findings.

Response: We have now added more details to understand model behavior and performance. This includes now a detailed sensitivity test that helps to understand how the resolution of N cycle affects N2O emissions. Further we include now time series of N2O emissions against data for a suite of sites. All these data show that resolving and predicting N2O emission is a challenge for any model.

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Responses to A.F. Bouwman (Referee)

Many thanks for reviewing our manuscript and your suggestions are very help in improving our study. Please find our responses below.

-This MS presents a module for simulating N2O fluxes at the global scale based on equations for denitrification and nitrification and considering N2O and NOx as fractions of the nitrogen that is processed. Most model elements were borrowed from other models. I have a number of serious problems with this MS:

Response: Thank you for taking time reviewing our paper.

-The model description in Appendix A is not complete as the units are not provided.

Response: Agree. Appendix A is rewritten and moved to sect. 2.1.2 Soil N2O emission. We have further added more explanation on the overall plant-soil nitrogen cycle. We believe we caught all instances where we missed the units.

-It is not clear how model calculations at a resolution of 3.75 by 2.5 degrees can be meaningful, since all data such as weather, soil and vegetation are kind of aggregates for that resolution, and how can this be compared with point measurements.

Response: We agree with the reviewer that evaluations of global simulations against point measurements result in scale mismatches. Soil N2O fluxes are highly variable even at the scale of a single stand. Currently, a higher resolution that is feasible for global simulation (e.g. 0. 5 by 0. 5 degrees) is still much larger than the area where field measurements take place, and benchmarking global simulations against field measurements still faces scale mismatches. In this way, any model data comparison is incomplete. In the original manuscript, we focus on capturing the average of annual mean emissions across different observations instead of one-by-one comparison. This overall average emission aggregates measurements and provides some information on model performance. We make reference to scale mismatches in the text. We further amend our manuscript with comparison against single points (although these points still represent grid-cell averages). We believe this is still informative, for example, our analysis

shows that, while we somewhat capture the means of the fluxes across sites. However, our model has trouble simulating peak emissions. Further studies may elucidate whether a better representation of these hot moments are more sensitive to processes emitted (freeze-thaw cycles, pulses), or the aggregation of vegetation and climate.

-*With this spatial resolution, the time step is 30 minutes, but the authors provide annual and seasonal numbers only. It is probably more interesting to compare the model results with temporal distributions from field measurements. This could be done for a number of test sites in a variety of climate and soil conditions. If not available, perhaps seasonal estimates from experimental sites could be used to validate the model.*

Response: We have now added monthly and daily measurements to sect. 2.3 and 3.3, which show, that the model agrees with the general trends in N2O emission, but measured emission peak are not realized in the model.

-*To assess model quality, it is much more interesting to analyze the functioning of the soil-plant nitrogen cycle. How is denitrification compared to field measurements, and leaching, plant uptake, ammonia volatilization, etc. If the large flows in the system are correct, the authors will also be more confident about the small fluxes like N2O and NOx.*

Response: Assessing model quality is a challenge for all models, and several reviewers have pointed out the effect of the larger N cycle on nitrification, denitrification and associated N2O fluxes. We now present an extensive sensitivity analysis that evaluates N2O fluxes in response to variation the relative strength of plant uptake, the overall supply of N to mineralization (by way of modifying biological N fixation), and by specific parameters that deal with nitrification and denitrification. We discuss the hierarchical system of plants and soils N demand are strong sinks for ammonium and nitrate under limiting condition, resulting in leaching and denitrification very small amounts. Excess nitrogen (if plants and soils have sufficient N) leads to leaching and denitrification. There, the partition coefficients and the parameters that determine the fate of the excess N become very important. The sensitivity analysis targets exactly that question. We further point out, that we do not invent a new model but put existing formulation (and parameter choices) in a new model. Currently, we do not have large scale observation data such as denitrification and ammonia volatilization in relative pristine

ecosystems to support the benchmarking of the global model. Detailed analyses addressing this are now in sect. 2.2.3, 3.4 and the discussion part of the revised manuscript.

-Finally, a true sensitivity analysis will also show what the major variables and parameters are. For example, the N2O and NOx fractions will probably pop up as important coefficients.

Response: We add a series of sensitivity tests with regard to plant N uptake, nitrification rates, denitrification rates and the fraction of N2O lost from net nitrification in sect. 2.2.3 and sect.3.4 of the revised manuscript, which shows effects of the larger N cycle on the availability of N for nitrification and denitrification as well as direct parameter uncertainty of the added module. We found that the fraction of N2O lost from net nitrification is the most sensitive parameter.However, this fraction is very uncertain based on limited field or laboratory studies. Goodroad and Keeney (1984) suggested a value of 0.1-0.2% , while Khalil et al. (2004) reported a range of 0.16%-1.48% depending on the O_2 concentration. We applied a value of contant 0.4% in the default run which embraces large uncertainties in our modelled results.

-I fully agree with one of the other reviewers who states that this work is not ready for publication, and I also agree that perhaps Geoscientific Model Development is a more appropriate journal for submitting a revised MS.

Response: Thanks again for the helpful suggestions. We carefully considered the possibility for Geoscientific Model Development. As pointed out in the response to Beni Stocker's review, we highlight that this is not a new development, but the addition of an existing nitrification/denitrification module to LM3VN. We would like to emphasize that the basic goal of this paper is thus not presenting the new module per se, but how it performs in the context of the larger plant-soil N cycle. The reviews have helped to sharpen this focus, which we think makes the manuscript a nice fit for Biogeosciences.

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5 Biochemistry, 36, 687-699, doi:10.1016/j.soilbio.2004.01.004, 2004.

6

Responses to Anonymous Referee #3

The authors added a new soil N2O emissions module to the dynamic global land model LM3V-N, and tested its sensitivity to soil moisture regime, as well as its responses to elevated CO2 and temperature. However, I am not sure what the main objective of the paper is – whether this was mainly a model development paper or whether they wanted to conduct different sensitivity analyses. As noted by the other two reviewers, I think this paper needs major revisions before it can be published. In my opinion, the most important is to: 1) include more analyses instead of the speculations presented in its current status; and 2) highlight the original contributions in this paper, specifically illustrating what is different from what already has been published in Xu-Ri et al., 2012.

Response: In the new manuscript we hope that we clarify that we are building on existing nitrification/denitrification modules and discuss how this implementation bears out in this specific coupled carbon- nitrogen cycle model. As pointed out by Beni Stocker, these processes are subject to how the larger plant-soil cycle is implemented. To achieve this goal, we added a set of new analysis and tested the response of N_2O emissions to assumptions related to N availability for nitrification-denitrification through altering biological N fixation fluxes, limiting dissolved organic N and fire volatilization N losses and changing plant N uptake strength. The corresponding analyses are in presented in sect. 2.2.3 and sect. 3.4 (***Sensitivity to N cycling processes and parameterization***) and the discussion in the revised manuscript. Our modeled response to CO_2 fertilization is different from Xu-Ri et al. (2012). Xu-Ri et al. (2012) suggests a positive response globally or from tropical forest based on histroical simulations and combining the interaction with climate change, while we argue for a negative response from tropical forest in the first three decades of imposing a doubling of atmospheric CO_2 (568 ppm).

I first list some major concerns, followed by minor comments.

Major points:

The authors argue in the abstract ln. 7-9 on p. 3102 that "[t]he model was capable of reproducing the average of cross-site observed annual mean emissions, although differences remained across individual sites if stand-level measurements were representative of gridcell

emissions." It is not obvious how they concluded that the model was indeed capable of reproducing the observed emissions. From the Figure 3, it is also not clear if the model is capable or not.

Response: We now add more comparison against data, specifically we also compare time series for a suite of site against the model. This allows us to discuss the model results in much more detail. For example, we show and discuss in the text, that we do not capture the entire breadth of N2O emissions across sites, and also within particular sites. Our abstract reads now "Results extracted from the corresponding gridcell (without site-specific forcing data) were comparable with the average of cross-site observed annual mean emissions, although differences remained across individual sites if stand-level measurements were representative of gridcell emissions.". While mismatches uncover model deficiencies, a point by point evaluation also bears the problem of scale mismatches, and issue raised by other reviewers and discussed in the text.

I would expect to see more rigorous model-obs comparisons, if this is a model development/validation paper. As the second reviewer suggested, I would also like to see hourly/monthly comparisons at multiple sites, and I find it odd that the model is run at "an annual time step" as they state on ln. 8-9, p. 3113.

Why don't they get the annual average from their half-hourly simulation?

Response: The model is run with the fastest time-step of half an hour. The results state on ln. 8-9, p. 3113 (from original manuscript) is the annual average from the half-hourly simulation. To clarify, we reworded to "Modelled N_2O emissions capture the average of cross-site observed annual mean emissions" (ln. 4-6, p. 16, revised manuscript). We further added comparison based on monthly and daily site measurements to sect. 2.3 and 3.3 of the revised manuscript..

Also, I recommend that they at least add their modeled values in Table B1 as well, so that the reader can directly compare their modeled values to the observations.

Response: We followed the suggestion:. Table B1 is moved to Table A1, and modeled values are added.

With regard to soil moisture, why does Figure 3 use different methods for the different data sets? I understand that there are three methods that the authors used for each of the three different data sets but it does not make much sense to do a model-obs comparison in a panel, using method 3 for part a and method 2 for parts b and c. Why not use one of the methods for all parts? If the authors agree that soil moisture values larger than 0.6 are not reasonable, what about the validity of the maximum water method that leads to a global mean WFPS higher than 0.6 (Figure 4)?

Response: We have three methods for each of the three different data sets. LM3V-SM does not allow soil water to accumulate beyond field capacity. Meanwhile, the other two data sets (NOAH-SM and ERA-SM) are products to emulate observed soil moisture where soil water can transiently be stored above field capacity. Based on our understanding of these soil water data sets, we believe WFPS is more accurately represented by method 3 for LM3V-SM (part a), and by method 2 for NOAH-SM (part b) as well as ERA-SM (part c). Therefore, we use different method for different soil moisture data set. WFPS higher than 0.6 are generated by the two external soil moisture data sets (NOAH and ERA) through the maximum method, which we mention in the text is less appropriate to use for these data, nevertheless, they provide useful information in terms of the sensitivity to the soil moisture and its parameterization.

I also found that there are some statements in this paper that should be better justified. First, on ln. 5, p. 3115 authors state that "[t]he negative impacts (reduced N2O flux), which are also reported from manipulative experiments, are likely from increased plant N and immobilization demand under CO2 fertilization, reducing N availability for nitrifiers and denitrifiers" but is this what they see in the model? I believe they can also draw a similar graph, illustrating plant N and immobilization rate in time-series to see if this is indeed the response they are seeing in the model. The same goes for the positive impacts.

I think it is important to see if the litter production and soil moisture have been increased, as well as stomatal conductance and leaf transpiration reduction, as they imply in the paper.

Response: This is a great suggestion. We inserted a new figure (Figure 8) into the revised manuscript. The figure compares the global mean litter pool size, plant nitrogen uptake rate, transpiration and soil water content in the root zone between simulations without and with CO_2 fertilization. Averages of global means over 100 years show an increase of plant nitrogen uptake

rate, litter pool size and soil water content, and a decrease of transpiration due to CO_2 fertilization effect.

I'm not sure I understand the reasoning behind the statement on ln. 14-16, p. 3118: "Patterns of seasonality, and the correlates between N2O emissions vs. temperature and soil moisture suggest that moisture is the dominant driver of N2O emission in tropical regions and soil temperature critical elsewhere." What does "dominant" mean in this case? I think that in order to make such a statement, one needs to show the impact of different variables that are important and how that affects their N2O emissions.

Response: According to other reviewer's suggestion, the correlation analysis is removed from the manuscript.

The authors write on p. 3104 that "[s]imulations with LPJ-DyN and O–CN demonstrated a positive response of N2O emissions to historical warming and a negative response to historical CO2 increase, globally. This negative CO2 response seems to be in disagreement with one meta-analysis of manipulative field experiments showing an increase in N2O emissions at elevated levels of CO2 (Zaehle et al., 2011; Xu et al., 2012; van Groenigen et al., 2011). The discrepancy in response to global change factors needs to be addressed both in models and in the interpretation of manipulative field experiments." It seems that authors are misinterpreting the work of Xu-Ri et al. (2012) (which authors write as Xu et al, (2012)). Xu-Ri et al. (2012) states that "[i]ncreasing CO2 generally enhanced the N2O emission in tropical and temperate moist forests, whilst reducing the N2O emission in some other regions (Fig. 6)," which is essentially the same as the argument made in the current paper. I think it would be helpful if the authors could clarify what it is that they are arguing that is different from the conclusions in the Xu-Ri et al. (2012), as this was not obvious to me.

Response: Fig. 6 of Xu-Ri et al., (2012) displays the simulated global 20th century trends of annual N_2O emission in simulations with (a) CO_2 and climate change and (b) fixed CO_2 concentration. Xu-Ri et al. (2012) states "in many tropical regions, CO_2 and climate change combined synergistically to increase N_2O emission", based on their Fig .6. However, the effect of CO_2 alone cannot be derived from their Fig. 6. As further illustrated in their Fig. 7, CO_2 plus interaction with climate result in a positive response of global N_2O emissions, but historical

CO_2 change alone (single factor) causes a slight decrease in historical N_2O emissions. We agree our interpretation of their result is inaccurate without explicitly state whether it is CO_2 effect alone or CO_2 plus interaction with climate. In response to this and another reviewer's suggestion, we rewrote this part as: „Simulations with O-CN demonstrated a positive response of N_2O emissions to historical warming and a negative response to historical CO_2 increase, globally. While CO_2 and interaction with climate change resulted in an increase in historical and future N_2O emissions from LPJ-DyN(Xu-Ri et al., 2012) and its application (Stocker et al. 2013), respectively, historical CO_2 change alone (single factor, from Fig. 7 of Xu-Ri et al., (2012)) caused a slight decrease in historical N_2O emissions."

We do not think our argument for CO_2 fertilization response is the same as Xu-Ri et al., (2012). Xu-Ri et al., (2012) argues for a positive response from tropical forest based on histroical simulations and combining the interaction with climate change, while we produced a negative response from tropical forest in the first three decades of imposing a doubling of atmospheric CO2 (568 ppm). The negative response from tropical forests is the major cause of the global negative responses to CO_2 fertilization. While Xu-Ri et al., (2012) conducted historical simulations, we focus on step changes of CO_2 that mimic most of the field experiment of CO_2 fertilization (e.g. FACE).

Minor comments:

I am a bit confused about the Figure 1. The MEI values (http://www.esrl.noaa.gov/psd/enso/mei/table.html) are higher than 0.6 on several occasions between 1975 and 1980 (1976 Jun-Oct, 1977 Jun-1978 Mar, 1979 Jul-1980 Jul) and yet, this figure is only showing a one gray zone during that period. Also, it is unclear which WFPS method was used for this calculation. It would be helpful if they showed the range in interannual emissions, based on the 3 different methods and datasets they used. The same goes for Figure 2.

Response: Agree. The revised Figure 1displays three sets of annual global N_2O emissions corresponding to three soil moisture datasets. We used the average of 12 monthly values to represent MEI of a year. Grey zones indicate the years with mean MEI greater than 0.6. And grey areas do not incorporate any sub-annual information.

Are RNOx:N2O and RN2:N2O values calculated at every time step for every grid cell? Or how does it work?

Response: Yes. RNOx:N2O and RN2:N2O values are calculated at every time step for every grid cell, which we now expliclitly mention in the revised manuscript.

L. 4, P. 3102 – typo "reponses"

L. 19, p. 3109 – typo "equalibrium".

l. 1, p. 3117 – typo "exsit"

l. 5, p. 3117 – "constraint" to "constrain"

l.29, p. 3117 – typo "oringinal" l. 8, p. 3119 – typo "aboitic".

l. 13, p. 3119 – typo "unstand"

l. 9, p. 3120, typo – "speicies"

Response: Thank you for catching those typos.

Brief List of Relevant Changes

According to the reviewer's suggestion, we made the following major changes:

1) We removed the appendix, and inserted the relevant equations that drive the dynamics of the ammonium and nitrate pool into the method section.
2) We conducted a series of sensitivity tests, that include both parameters directly associated with the fast turnover pools of ammonium and nitrate (and thus nitrification and denitrification rates), and processes that affect the larger plant-soil N cycle. The parameters include maximum uptake of plant roots, maximum nitrification rate, maximum denitrification reate, half saturation constants for C availability and nitrate during denitrification and the fraction of nitrification lost as N_2O. The sensitivity to the larger plant-soil N cycle includes a) a prescribed rate of biological N fixation instead of the dynamic changes formulated in LM3V-N, changes in the fraction of N volatilized during fire, and a parameter that affects production and losses of dissolved organic N. To that end, we provide a new figure (figure 7).
3) We provide site-specific evaluations, of N2O fluxes, and how simulated N2O at these sites responds to the different treatments of soil moisture in the model. These evaluations resulted in 2 new figures (Figures 5 and 6).
4) According to the reviewer's suggestion, we removed the old figure 2 that showed correlations of N2O fluxes with other variables.
5) We further created a new figure (figure 9) that shows the response of selected state variables and N fluxes in response to elevated CO_2. This is in response to the request of reviewer 3, to discuss how the larger plant-soil N cycle shapes the response of N_2O emission to a doubling of CO_2.
6) Following reviewer's comments, we expanded the discussion to accommodate comments regarding how the paper is interesting for the readers of Biogeoscience. We specifically highlight, that we build on earlier, established formulations of nitrification/denitrification and we show, how the implementation of the larger plant-soil N cycle affects the modelling of the soil ammonium and nitrate availability – and thus N2O emissions.

Marked up version of the manuscript changes

Abstract

Nitrous oxide (N_2O) is an important greenhouse gas that also contributes to the depletion of stratospheric ozone. ~~With~~Due to its high temporal and spatial heterogeneity, a quantitative understanding of terrestrial N_2O emission, its variabilities and ~~reponses~~responses to climate change is challenging. We added a soil N_2O emission module to the dynamic global land model LM3V-N, and tested its sensitivity to ~~soil moisture regime and responses to elevated CO_2 and temperature. The model was capable of reproducing~~mechanisms that affect the level of mineral N in soil such as plant N uptake, biological N fixation, amount of volatilzed N redeposited after fire, and nitrification. We further tested the relationship between N_2O emission and soil moisture, and finally assessed responses to elevated CO_2 and temperature. Results extracted from the corresponding gridcell (without site-specific forcing data) were comparable with the average of cross-site observed annual mean emissions, although differences remained across individual sites if stand-level measurements were representative of gridcell emissions. Processes, such as plant N uptake and N loss through fire volatilization, that regulate N availability for nitrification-denitrification have strong controls on N_2O fluxes in addition to the parameterization of N_2O loss through nitrification and denitrification. Modelled N_2O fluxes were highly sensitive to water filled pore space (WFPS), with a global sensitivity of approximately 0.25 TgN per year per 0.01 change in WFPS. We found that the global response of N_2O emission to CO_2 fertilization was largely determined by the response of tropical emissions~~, whereas~~ with reduced N_2O fluxes in the first few decades and increases afterwards. The initial reduction was linked to N limitation under higher CO_2 level, and was alleviated through feedbacks such as biological N fixation. The extratropical response was weaker and ~~different~~generally positive, highlighting the need to expand field studies in tropical ecosystems. Warming generally enhanced N_2O efflux, and the enhancement was greatly dampened when combined with elevated CO_2, although CO_2 alone had a small effect. Our analysis suggests caution when extrapolation from current field CO_2 enrichment and warming studies to the global scale.

1 Introduction

Nitrous oxide (N_2O) is a major reactant in depleting stratospheric ozone as well as an important greenhouse gas ~~(Ravishankara et al., 2009~~(Ravishankara et al., 2009;Butterbach-Bahl et al., 2013;Ciais et al., 2013). With a global warming potential of 298 times more (per unit mass) than that of carbon dioxide (CO_2) over a 100-year period (Forster et al., 2007), the contributions of N_2O emissions to global radiative forcing and climate change are of critical concern ~~(Zaehle and Dalmonech, 2011)~~(Zaehle and Dalmonech, 2011). The concentration of atmospheric N_2O has been increasing considerably since the industrial revolution with a linear rate of 0.73±0.03 ppb yr^{-1} over the last three decades (Ciais et al., 2013). Although applications of synthetic fertilizer and manure during agriculture intensification have been identified as the major causes of this increase which has resulted in an increase of the radiative forcing by 0.125W m^{-2} ~~(Davidson, 2009;~~(Davidson, 2009;Zaehle and Dalmonech, 2011~~);~~ Zaehle et al., 2011), nonagricultural (natural) soil is still an important N_2O source ~~that is comparable to the combined anthropogenic emissions~~ (Ciais et al., 2013;Syakila and Kroeze, 2011). N_2O fluxes from nonagricultural soils are highly heterogeneous, which limits our ability to estimate and predict global scale budget, and quantify its response to global environmental changes (Butterbach-Bahl et al., 2013;Ciais et al., 2013).

Most of the N_2O fluxes from soil are produced by microbial nitrification and denitrification (Braker and Conrad, 2011;Syakila and Kroeze, 2011). Nitrification is an aerobic process that oxidizes ammonium (NH_4^+) to nitrate (NO_3^-), during which some N is lost as N_2O. Denitrification reduces nitrate or nitrite to gaseous N (i.e. NO_x, N_2O and N_2), a process that is fostered under anaerobic conditions. N_2O is generated in intermediary steps during denitrification and a small portion can escape from soil before further reduction to N_2 takes place. Soil texture, soil NH_4^+, soil water filled pore space (WFPS), mineralization rate, soil pH, and soil temperature are well-known regulators of nitrification N_2O fluxes ~~(Parton et al., 1996;Li et al., 2000;Parton et al., 2001)~~(Parton et al., 1996;Li et al., 2000;Parton et al., 2001). Denitrification and associated N_2O emissions depend primarily on carbon supply, the redox potential and soil NO_3^- (Firestone and Davidson, 1989;~~Parton et al., 1996)~~Parton et al., 1996). Soil moisture has a ~~particular~~particularly strong impact ~~(Galloway et al., 2003;Schlesinger, 2009)~~(Galloway et al., 2003;Schlesinger, 2009) as it influences nitrification and denitrification

rates through its regulations on substrate availability and soil redox potential (as oxgyen diffusion proceeds at much slower rate in water filled than in air filled pore space), thereby also controlling the partitioning among various denitrification products (i.e. NO_x, N_2O and N_2) (Firestone and Davidson, 1989;~~Parton et al., 2001)~~Parton et al., 2001). Although emissions are known to be sensitive to soil moisture, quantitative understanding of its role in terrestrial N_2O fluxes and variability is limited (Ciais et al., 2013).

At regional to global scale, the application of the "hole-in-pipe" concept (Firestone and Davidson, 1989) in the CASA biosphere model pioneered one of the earliest process-based estimation of natural soil N_2O fluxes. The model calculated the sum of NO, N_2O and N_2 fluxes as a constant portion of gross mineralized N, and the relative ratios of N trace gases (NO_x:N_2O:N_2) as a function of soil moisture ~~(Potter et al., 1996)~~(Potter et al., 1996). While the early models of nitrification and denitrification are primarily conceptual driven, recent global N_2O models combine advancements in global dynamic land models with more detailed processes, including microbial dynamics. ~~Xu and Prentice (2008)~~Xu-Ri and Prentice (2008) simplified nitrification and denitrification modules from DNDC (i.e., DeNitrification-DeComposition) ~~(Li et al., 1992;Li et al., 2000)~~(Li et al., 1992;Li et al., 2000) in their global scale dynamic N scheme (DyN) and incorporated DyN into the LPJ dynamic global vegetation model. In the DNDC approach, nitrification and denitrification were allowed to happen simultaneously in aerobic and anaerobic microsites. Zaehle et al. (2011) incorporated a nitrification-denitrification scheme into the O-CN land model following largely the LPJ-DyN with minor modifications and additions of the effects of soil pH and chemo-denitrification that originated from DNDC ~~(Li et al., 2000).~~(Li et al., 2000). Stocker et al. (2013) embeded the LPJ-DyN approach into an Earth System Model and investigated the feedbacks of N_2O emissions, together with CO_2 and CH_4, to climate. Compared to LPJ-DyN approach, ~~Saikawa et al. (2013)~~Saikawa et al. (2013) retained the explicit simulation of nitrifying and denitrifying bacteria from DNDC in their CLMCN-N2O module based on CLM V3.5 land model. Simulations with ~~LPJ-DyN and~~ O-CN demonstrated a positive response of N_2O emissions to historical warming and a negative response to historical CO_2 increase, globally. ~~This~~While CO_2 and interaction with climate change resulted in an increase in historical and future N_2O emissions from LPJ-DyN (Xu-Ri et al., 2012) and its application in LPX-Bern (Stocker et al., 2013), respectively, historical CO_2 change alone, i.e. single factor of Xu-Ri et al., (2012), caused a slight decrease in historical N_2O emissions. The negative CO_2 response seems to be

in disagreement with one meta-analysis of manipulative field experiments showing an increase in N_2O emissions at elevated levels of CO_2 (Zaehle et al., 2011;~~Xu et al., 2012~~Xu-Ri et al., 2012;van Groenigen et al., 2011). The discrepancy in response to global change factors needs to be addressed both in models and in the interpretation of manipulative field experiments.

Here we add a N_2O gas emission module to LM3V-N, a land model developed at the Gephysical Fluid Dynamics Laboratory (GFDL). In this paper, we will first briefly introduce LM3V-N and describe the added N_2O emission module. We then subject the model to historic changes in CO_2, N deposition, and recent climate change to infer natural N_2O emissions in the past few decades. We test the model's sensitivity to soil water regime, by addressing the parameterization of soil WFPS, and by ~~data overriding of two different soil moisture reanalysis products. We then~~replacing the model soil moisture with two different soil moisture reanalysis products. We also conduct sensitivity tests with regard to the general N cycling and parameterization of N_2O emissions. Since we build largely on existing parameterization of nitrification-denitrification processes, our focus relies on the evaluation of these processes if tranferred to a different model. Finally, we subject the model to step changes in atmospheric CO_2 and temperature to understand modelled reponses to CO_2 fertilization/climate change.

2 Methods

2.1 Model description

LM3V is capable of simulating ecosystem dynamics and exchange of CO_2, water and energy between land and atmosphere with the fastest time step of 30 minutes ~~(Shevliakova et al., 2009).~~(Shevliakova et al., 2009). LM3V-N expands the LM3V land model with a prognostic N cycle ~~(Gerber et al., 2010)~~(Gerber et al., 2010), and includes five plant functional types (PFTs):C3 and C4 grasses, tropical, temperate deciduous and cold evergreen trees. Each PFT has five vegetation C pools (~~leaves~~leaf, fine ~~roots~~root, sapwood, labile, and wood), two litter and two soil organic C pools and their corresponding N pools based on the specific C:N ratios. Photosynthesis is coupled with stomatal conductance on the basis of the Collatz et al., (1991,1992) simplification of the Farquhar scheme (Farquhar et al., 1980). ~~N enters the ecosystem through atmospheric N deposition and biological N fixation (BNF). BNF in LM3V-N is dynamically simulated on the basis of plant N availability, demand and light condition.~~ Soil hydrology in LM3V follows partly on Land Dynamics (LaD) with further improvements

(Shevliakova et al., 2009;Milly and Shmakin, 2002;Milly et al., 2014). N enters the ecosystem through atmospheric N deposition and biological N fixation (BNF), losses via fire and leaching of dissolved organic N (DON) as well as mineral N. Major characteristics of LM3V-N include the following 5 aspects, and details are available in Gerber et al. (2010).

2.1.1 Main characteristic of LM3V-N

2.1.1.1 C-N coupling in vegetation

We briefly describe the larger plant-soil N cycle and how it links to mineral N (ammonium and nitrate). Details are described in Gerber et al. (2010). Plants adjust their uptake of C and N to maintain their tissue specific C:N ratios, which are PFT-dependent constants. Instead of varying C:N ratios in tissues, short-term asynchronies in C and N assimilations or temporary imbalances in stoichiometry are buffered by additional N storage pool (S) in which N is allowed to accumulate once plant N demand is satisfied. The optimum storage size S_{target} is based on tissue turnover $Q_{N,liv}$,

$$S_{target} = t_h Q_{N,liv} \quad (1)$$

where t_h is the time span that buffer plant N losses (currently set as 1 year). Plant N status (x) is defined as the fraction of the actual N storage compared to the target storage: $x = S/S_{target}$. Consequently, N constraints on photosynthesis and soil N assimilation are based on plant N status:

$$A_{g,N} = A_{g,pot}(1 - e^{-x\varphi}) \quad (2)$$

$$U_{N,P} = U_{N,P,pot} * \begin{cases} 1 & if\ S < S_{target} \\ 0 & else \end{cases} \quad (3)$$

where $A_{g,N}$ indicates N constrained rate of gross photosynthesis (μmolC m^{-2} s^{-1}) and $A_{g,pot}$ corresponds to the potential photosynthetic rate without N limitation. The parameter φ mimics the metabolic deficiency as plant N decreases. $U_{N,P,pot}$ is the potential inorganic N uptake rate from soil available ammonium and nitrate pools. The actual inorganic N uptake rate ($U_{N,P}$) operates at its potential and drops to zero when N storage (S) reaches its target size.

2.1.1.2 Soil C-N interactions in organic matter decomposition

Organic matter decomposition is based on a modified CENTURY approach (Bolker et al., 1998), and amended with ~~formulation of N dependent C and N mineralization rates .~~

formulations of N dependent C and N mineralization rates. N can both trigger the decomposition of "light" organic matter and stabilize C in "heavy" organic matter in LM3V-N. Sustained positive effect of available N on litter decomposition relies on the persistence of microbial N limitation during decomposition, which is implemented through the combination of available N supply to microbial organisms and their respiration rate. Further, LM3V-N incorporates the negative effects of N on recalcitrant organic matter decomposition through increasing the fraction of C and N fluxes into the recalcitrant pool. Formation of a slow decomposable organic matter pool leads to immobilization of ammonium and nitrate to satisfy the fixed carbon to nitrogen ratio of this pool.

2.1.1.3 Competing sinks of available N

The fate of soil mineral N (i.e. ammonium and nitrate) depends on the relative strength of the competing sinks, with the broad hierarchy ~~order~~ of sorption > soil immobilization > plant uptake > leaching/denitrification. Denitrification thus far ~~is~~has been lumped with leaching losses and summed into a generic N loss term. ~~Over~~Sorption/desorption buffers available N and is assumed to have the ~~long term,~~highest priority and be at steady state in each model time step. N immobilization into organic matter occurs during transfers among litter and soil organic matter pools. Leaching losses of ~~N from fire and dissolved organic nitrogen (DON)~~ available N are ~~critical factors limiting~~ simulated on the ~~ecosystem N accumulation and maintaining N limitation in LM3V~~ basis of drainage rate. Plant uptake of mineral N ~~(Gerber et al., 2010;Gerber et al., 2013). Soil hydrology in LM3V follows partly on Land Dynamics (LaD) with further improvements (Shevliakova et al., 2009;Milly and Shmakin, 2002;Milly et al., 2014).~~is a combination of both active and passive processes. The active uptake is modeled as a Monod function, and the passive transport is a function of available N and plant transpiration.

~~Here, we~~ $$U_{N,P,pot,i} = \frac{v_{max} C_r N_{i,av}}{h_s (k_{p,1/2} + [N_{av}])} + [N_{av}] Q_{W,T} \quad (4)$$

where v_{max} (yr^{-1} kgC^{-1}) stands for the maximum uptake rate per unit root mass C_r, h_s is soil depth, $k_{p,1/2}$ is the half saturation constant, and $Q_{W,T}$ represents the transpiration flux of water. Potential uptake and thus effective removal of available N occurs if plants are N limited (see Equation 3).

2.1.1.4 N losses from organic pools

Over the long term, N losses via fire and DON are critical factors limiting ecosystem N accumulation and maintaining N limitation in LM3V-N (Gerber et al., 2010; Thomas et al., 2015). N volatilized from fire is approximated as a function of C released from fire, stoichiometric ratio of burned tissues and reduced by a global retention factor representing the fraction of N that is retained as ash (*ash_fraction*, currently set as 0.45). DON leaching is linked to hydrologic losses of dissolved organic matter (L_{DOM}) and its C:N ratio. In turn L_{DOM} is based on drainage rate ($Q_{W,D}$) and a buffer or sorption parameter b_{DOM} (currently set as 20).

$$L_{DOM} = \frac{Q_{W,D}}{h_s b_{DOM}} DOM \quad (5)$$

where *DOM* is the amount of dissolve organic matter in the soil column. Soil depth (h_s) is used to convert DOM unit to concentration (in unit of kgC m^{-3}). Production of DOM (in unit of kgC m^{-2}) is assumed to be proportional to the decomposition flux of the structural litter and soil water content. Both, losses via fire and via DOM are losses from a plant-unavailable pool (Thomas et al., 2015), and have the potential to increase or maintain N limitation over longer timescales, and consequently reduce N available for N_2O production through sustained and strong plant N uptake (see Equations 2-4).

2.1.1.5 Biological nitrogen fixation (BNF)

BNF in LM3V-N is dynamically simulated on the basis of plant N availability, N demand and light condition. BNF increases if plant N requirements are not met by uptake. The rate of up-regulation is swift for tropical trees but constrained by light penetrating the canopy for other PFTs, mimicking the higher light requirements for new recruits that possibly can convert atmospheric N_2 into plant available forms. In turn, sufficient N uptake reduces BNF. The BNF parameterization thus creates a negative feedback, where high plant available N and thus the potential for denitrification is counteracted with reduction of N input into the plant-soil system. This explicit negative feedback is different to other models where BNF is parameterized based on NPP (Thornton et al., 2007), or transpiration (Zaehle and Friend, 2010).

2.1.2 Soil N_2O emission

LM3V-N assumes that nitrification is linearly scaled to ammonium content, and modified by soil temperature and soil moisture. Gaseous losses so far were not differentiated from hydrological leaching. We add a soil nitrification-denitrification module which accounts for N gaseous losses from NH_3 volatilization, nitrification and denitrification. The nitrification-

denitrification scheme implemented here combines features from both the DNDC model ~~(Li et al., 1992;Li et al., 2000)~~(Li et al., 1992;Li et al., 2000) and the CENTURY/DAYCENT ~~(Parton et al., 1996;Parton et al., 2001~~(Parton et al., 1996;Parton et al., 2001;Del Grosso et al., 2000). In this part, we provide details on the nitrification-denitrification module which explicitly simulates N gaseous losses from nitrification and denitrification, as well as other process modifications compared to the original LM3V-N.

2.1.2.1 Nitrification-Denitrification

Transformation among mineral N species (ammonium and nitrate) occurs mainly through two microbial pathways: nitrification and denitrification. Although ongoing debate exists in whether nitrification rates may be well described by bulk soil ammonium concentration or soil N turnover rate (Parton et al., 1996;Zaehle and Dalmonech, 2011), we adopt the donor controlled scheme (ammonium concentration). In additon to substrate, soil texture, soil water filled pore space (WFPS, the fraction of soil pore space filled with water), and soil temperature are all well known regulators of nitrification. As a first order approximation, nitrification rate (*N*, in unit, kgN m^{-2} $year^{-1}$) is simulated as a function of soil temperature, NH_4^+ availability and WFPS,

$$N = k_n f_n(T) f_n(WFPS) \frac{N_{NH_4^+}}{b_{N,NH_4^+}} \quad (6)$$

where k_n is the optimum nitrification rate (11000 $year^{-1}$, the same as in LM3V-N) (Gerber et al., 2010); $N_{NH_4^+}$ is ammonium content (in unit, kgN m^{-2}); b_{N,NH_4^+} is the buffer or sorption parameter for NH_4^+ (unitless, 10 in LM3V-N) (Gerber et al., 2010); $f_n(T)$ is the temperature response function following Li et al. (2000), with an optimum temperature for nitrification at 35°C; and $f_n(WFPS)$ is the soil water response function. The effect of WFPS on nitrification is texture dependent, with most of the reported optimum value around 0.6 (Parton et al., 1996;Linn and Doran, 1984). We adopt the empirical WFPS response function from Parton et al. (1996) with medium soil texture.

$$f_n(T) = \left(\frac{60 - Tsoil}{25.78}\right)^{3.503} \times e^{\frac{3.503 \times (Tsoil - 34.22)}{25.78}} \quad (7)$$

$$f_n(WFPS) = \left(\frac{WFPS - 1.27}{-0.67}\right)^{\frac{1.9028}{0.59988}} \times \left(\frac{WFPS - 0.0012}{0.59988}\right)^{2.84} \quad (8)$$

where *Tsoil* is the soil temperature in degree Celsius. ~~. Details of model formulation and implementation are given in Appendix A. Briefly, nitrification is treated as a donor (NH_4^+) controlled process which is further modified by soil moisture and temperature. Denitrification, a multiple step process that anaerobically reduces nitrate sequentially to the endproduct N_2, is simplified as a single process controlled by substrate NO_3^- (electron acceptor), labile C availability (electron donor), soil moisture and temperature. Heterotropical respiration (HR) is used as a surrogate for labile C availability, similar as Del Grosso et al. (2000) and Xu and Prentice (2008). WFPS plays a crucial role in the prediction of nitrification an denitrification, as it determines movement of dissolved molecules, and more importantly, puts strong constraints on movement of oxygen in soils, affecting the soil's redox potential. We therefore use WFPS to parameterize the soil's redox potential and substrate availability to nitrifying and denitrifying microbes.~~

Denitrification is controlled by substrate NO_3^- (electron acceptor), labile C availability (electron donor), soil moisture and temperature. Labile C availability is estimated by soil heterotrophic respiration (*HR*). Following LPJ-DyN (Xu-Ri and Prentice, 2008), denitrification is assumed to have a Q_{10} value of 2 when the soil temperature is between 15 and 25 °C. The soil moisture response function is adopted from Parton et al. (1996). Soil pH is reported to be an important indicator of chemodenitrification which occurs predominantly in acidic soils (pH<5) under conditions of high nitrite concentration (Li et al., 2000). However, its role for N_2O production is not well studied (Li et al., 2000) and we do not model the chemodenitrification explicitly.

$$D = k_d f_d(T) f_d(WFPS)\, f_g NO_3^- \quad (9)$$

$$\text{And } f_g = \frac{HR}{HR+K_C}\frac{NO_3^-}{NO_3^-+K_n} \quad (10)$$

$$NO_3^- = \frac{N_{NO_3^-}}{b_{NO_3^-}} \quad (11)$$

where *D* is the denitrification rate (in unit, kgN m^{-2} $year^{-1}$); k_d is the optimum denitrification rate (8750 $year^{-1}$); f_g mimics the impact of labile C availability and substrate (nitrate) on the growth of denitrifiers, adapted from Li et al. (2000); K_c and K_n are half-saturation constants taken from Li et al. (2000) (0.0017 and 0.0083 kgN m^{-2} respectively, assuming an effective soil depth of 0.1m); $b_{NO_3^-}$ is the buffer or sorption parameter for NO_3^- (unitless, 1 in LM3V-N)

(Gerber et al., 2010); $N_{NO_3^-}$ and NO_3^- are nitrate content before and after being buffered (in unit, kgN m^{-2}), respectively; and $f_d(T)$ and $f_d(WFPS)$ are empirical soil temperature and water reponse function for denitrification, adopted from Xu-Ri and Prentice (2008) and Parton et al. (1996), respectively.

$$f_d(T) = e^{308.56\times(\frac{1}{68.02}+\frac{1}{Tsoil+46.02})} \quad (12)$$

$$f_d(WFPS) = \frac{1.56}{12.0^{(\frac{16.0}{12.0(2.01\times WFPS)})}} \quad (13)$$

2.1.2.2 Gaseous partitions from nitrification-denitrification

N_2O is released as a byproduct from both nitrification and denitrification. The fraction of N_2O lost from net nitrification is uncertain (Li et al., 2000;Xu-Ri and Prentice, 2008). Here we set this fraction to be 0.4%, which is higher than Goodroad and Keeney (1984), but at the low end provided by Khalil et al. (2004). N_2O and NO_x emissions from nitrification are based on the NO_x: N_2O ratio ($R_{NOx:N2O}$) which is updated at every time step and for each grid cell. $R_{NOx:N2O}$ varies with relative gas diffusivity (D_r, the relative gas diffusivity in soil compared to air) (Parton et al., 2001), which is calculated from air filled porosity (*AFPS*, i.e., the portion of soil pore space that is filled by air) (Davidson and Trumbore, 1995)

$$R_{NOx:N2O} = 15.2 + \frac{35.5\times ATAN(0.68\times\pi\times(10\times D_r-1.68))}{\pi} \quad (14)$$

$$D_r = 0.209 \times AFPS^{\frac{4}{3}} \quad (15)$$

where ATAN stands for the trigonometric arctangent function; *AFPS* is the air filled porosity (1-WFPS), and π is the mathematical constant, approximately 3.14159.

During denitrification, the gaseous ratio between N_2 and N_2O ($R_{N2:N2O}$) is calculated following the empirical function derived by Del Grosso et al. (2000), which combines the effects of substrate (NO_3^-) to electron donor (*HR*, the proxy for labile C) ratio and WFPS. $R_{N2:N2O}$ is updated at every time step and for each grid cell.

$$R_{N2:N2O} = Fr(\frac{NO_3^-}{HR}) \cdot Fr(WFPS) \quad (16)$$

With

$$Fr\left(\frac{NO_3^-}{HR}\right) = \max(0.16 \times k, k \times e^{(-0.8\times\frac{NO_3^-}{HR})}) \quad (17)$$

$$Fr(WFPS) = \max(0.1, 0.015 \times WFPS - 0.32) \quad (18)$$

where k is a texture dependent parameter (Table 1) estimated from Del Grosso et al. (2000). k controls the maximum value of the function $Fr\left(\frac{NO_3^-}{HR}\right)$.

2.1.2.3 Other modified processes

To complete the N loss scheme in LM3V-N, we also added NH_3 volatilization into LM3V-N. NH_3 volatilization in soil results from the difference between the equilibrium NH_3 partial pressure in soil solution and that in the air. Dissolved NH_3 is regulated by ammonium concentration and pH. The net flux of NH_3 from soil to the atmosphere varies with soil NH_3, moisture, temperature, therefore

$$NH_3 = k_{nh} f(pH) f_{NH3}(T)(1 - WFPS)\frac{N_{NH_4^+}}{b_{N,NH_4^+}} \quad (19)$$

where NH_3 is the net ammonia volatilization flux (in unit, kgN m^{-2} year^{-1}); k_{nh} is the optimum ammonia volatilization rate (365 year^{-1}); $f(pH)$ is the pH factor and $f(T)$ is the temperature factor which are given by the following two equations:

$$f(pH) = e^{2\times(pH_{soil}-10)} \quad (20)$$

$$f_{NH3}(T) = \min\left(1, e^{308.56\times\left(\frac{1}{71.02}-\frac{1}{Tsoil+46.02}\right)}\right) \quad (21)$$

where pH_{soil} is the soil pH which is prescribed instead of simulated dynamically. $f(pH)$ and $f(T)$ follow largely on the NH_3 volatilization scheme implemented in the dynamic global vegetation model LPJ-DyN (Xu-Ri and Prentice, 2008).

2.2 Model experiments

2.2.1 Global hindcast with potential vegetation

To understand the model performance and compare with other models and observations, we conducted a hindcast simulation with potential vegetation. The model resolution was set to 3.75 degrees longitude by 2.5 degrees latitude. We forced the model with 3 hourly reanalysis weather data based on Sheffield et al. (2006)~~N_2O is released as a byproduct from both nitrification and denitrification. The fraction of N_2O lost from net nitrification is uncertain (Li et al., 2000;Xu and Prentice, 2008). Here we set this fraction to be 0.4%, which is higher than Goodroad and Keeney (1984), but at the low end provided by Khalil et al. (2004). Gaseous losses from denitrification is partitioned among N gaseous species (i.e. NO_x, N_2O and N_2) on the basis of~~

~~NO_x: N_2O ratio ($R_{NOx:N2O}$) (Parton et al., 2001) and N_2:N_2O ratio ($R_{N2:N2O}$) (Del Grosso et al., 2000). $R_{NOx:N2O}$ varies with gas diffusivity (Parton et al. 2001), which is estimated from air filled porosity (Davidson and Trumbore 1995). $R_{N2:N2O}$ combines the effects of substrate (NO_3^-) to electron donor ratio and WFPS (Del Grosso et al., 2000).~~

~~2.21.1 Model experiments~~

~~2.2.11.1.1 Global hindcast with potential vegetation~~

~~To understand the model performance and compare with other models and observations, we conducted a hindcast simulation with potential vegetation. The model resolution was set to 3.75 degrees longitude by 2.5 degrees latitude. We forced the model with 3 hourly reanalysis weather data based on Sheffield et al. (2006)~~. We used a 17 year recycled climate of 1948-1964 for the spin-up and simulation years prior to 1948. Atmospheric CO_2 concentration was prescribed with 284 ppm for model spin-up and based on ice core and atmospheric measurements for transient simulations ~~(Keeling et al., 2009)~~(Keeling et al., 2009). N deposition was set as natural background for simulations before 1850 (Dentener and Crutzen, 1994), and interpolated linearly between the natural background and a snapshot of contemporary (1995) deposition ~~(Dentener et al., 2006)~~(Dentener et al., 2006) for simulations after 1850. Soil pH was prescribed and derived from the Harmonized World Soil Database (HWSD) version 1.1, the same as NACP model driver data (Wei et al., 2014).

The model was spun up from bare ground without C-N interactions for the first 68 years and with C-N interactions for the following 1200 years to develop and equilibrate C and N stocks. ~~During~~To speedup the spin-up process, slow litter ~~C (N)~~ and ~~slow~~ soil C ~~(~~and N~~)~~ pools were set to the equilibrium values based on litterfall inputs and decomposition/leaching rates every 17 years. We determined the model to reach a quasi-equlibrium state by confirming the drift to be less than 0.03 PgC yr^{-1} for global C storage and 0.2 TgN yr^{-1} for global N storage. From this quasi equilibrium state, we initialized the global hindcast experiment starting from 1850 using the corresponding climatic forcings, CO_2 and N deposition data. In the following analysis, we will focus mostly on the last three decades (1970-2005~~) when most of the data are available.~~).

2.2.2 Sensitivity to soil water filled pore space (WFPS)

While LM3V-N carries a simplified hydrology, we bracketed effects of soil moisture by exploring the paremeterization of WFPS and by substituting the predicted soil moisture with 3-hourly re-analysis data. Levels of soil water (in ~~units~~unit kg m^{-2}) therefore stem from: (1) the simulated water content based on LM3V-N soil water module, hereafter LM3V-SM (2) the Global Land Data Assimilation System Version 2 with the land surface model NOAH 3.3 ~~(Rodell et al., 2004)~~(Rodell et al., 2004), hereafter NOAH-SM, and (3) the ERA Interim reanalysis dataset from European Center for Medium range Weather Forecasting (ECMWF) (Dee et al., 2011), hereafter ERA-SM. ~~Both of the later~~The latter two datasets integrate satellite and ground based obervations with land surface models. When overriding soil moisture, we linearly interpolated the 3 hourly data onto the 30 minutes model time step. In these simulations, we allowed soil C and N dynamics to vary according to different soil moisture datasets, but kept the model prediction of soil water to use for plant productivity and evapotranspiration.

Parameterization of the soil moisture effect on nitrification and denitrification are based on WFPS. LM3V-N uses the concept of plant available water, where ~~the maximum amount of~~ water ~~a soil can hold~~that is available to plants varies between the wilting point and field capacity. Water content above the available water capacity (i.e., the difference between field capacity and wilting point) leaves the soil immediately ~~(Milly and Shmakin, 2002)~~(Milly and Shmakin, 2002)~~,~~, and thus WFPS does not attain high values typically observed during denitrification. To ~~test~~explore the effect of WFPS – soil moisture relationship on N_2O emissions, we calcuated WFPS using three methods. Method 1 assumes WFPS is the ratio of available water and the available water capacity in the rooting zone. In ~~method~~Method 2 we assume, WFPS is the ratio of the water filled porosity and total porosity which is derived from bulk density (BD, in unit g cm^{-3}). BD was obtained from the Harmonized World Soil Database (HWSD) version 1.1 (Wei et al., 2014). The calculation is given by

$$WFPS = \frac{\frac{\theta}{h_r \times 0.001}}{1-\frac{BD}{2.65}} \quad \left(1-\frac{\frac{\theta}{\rho h_r}}{1-\frac{BD}{PD}}\right) \quad (22)$$

where θ (kg m^{-2}) is the root zone soil water; h_r (m) is the effective rooting depth of vegetation~~.~~; ρ is the density of water (1000 kg m^{-3}); and PD is the particle density of soil (2650 kg m^{-3}). Method 1 geerally leads ~~generally~~ to an overestimation of WFPS ~~with~~because the available

water capacity smaller than total pore space. In contrast, the use of ~~method~~Method 2 with LM3V-SM creates an underestimation since water is not allowed to accumulate beyond field capacity and misses high WFPS to which nitrification and denitrification are sensitive. Meanwhile, for NOAH-SM and ERA-SM data, Methods 2 ~~are~~is more close to the "real" WFPS~~.~~ and is the default method when using these data sets. In a third approach, which is also the default method with LM3V-SM that is applied in the global hindcast experiment ~~and~~, the ~~follwing~~subsequent elevated CO_2 and temperature responses experiment, and sensitivity tests with regard to N cycling, calculates WFPS as the average of the previous two methods.

For each soil moisture dataset (3 in total, 2 replacements and 1 simulated by LM3V-N), we calculated WFPS using three methods mentioned above. We conducted transient simulations with the nine different WFPSs (3 datasets × 3 methods) starting from the near equilibrium state obtained in the global hindcast experiment in 2.2.1. The use of less realistic Method for WFPS for each soil moisture driver (LM3V-SM, NOAH-SM and ERA-SM) offers insights of the sensitivity of N_2O emissions to soil moisture. The simulation procedure was the same as that in global hindcast experiment except for the WFPS. ERA-SM is only availabe starting from 1979, prior to which simulations were conducted with model default soil moisture (LM3V-SM). Results from ERA-SM were analyzed starting from 1982, leaving a short period for adjustment.

2.2.3 Sensitivity to N cycling processes and parameterization

N_2O emission is constraint by ecosystem availability of mineral N, which is linked to different N cycling processes in addtion to nitrification and denitrification processes. To test the sensitivity of modelled N_2O emission to the larger plant-soil N cycle, we conducted the following sensitivity analyses, in form of a one at a time perturbation. We replaced the dynamic BNF scheme with empirically reconstructed preindustrial fixation rates (Cleveland et al., 1999), removing the negative feedback between BNF and plant N availability. We further shut off N loss pathways through DON leaching and fire volatilization (with *ash_fraction* =1). We expect that these three modifications alleviate N limitation: Prescribed BNF may continuously add N beyond plant N demand. Further eliminating fire and DOM N losses leave loss pathways that have to pass the available N pool thereby opening the possibility of increasing gaseous losses. Further, removing these plant-unvailable pathways (Thomas et al., 2015) increases N retention and opens the possibility of alleviating N limitation. In addition, we modified key parameters related to general N cycling and N_2O emissions one-at-a-time. We multiplied several

parameters that directly affect ammonium and nitrate concentration or N_2O fluxes by 10 (x10) or 0.1 (x0.1), while kept other parameters as defaults. Those parameters control the active root N uptake rates (v_{max}), nitrification rate (k_n), denitrification rate (k_d, Kc,Kn) and the fraction of net nitrification lost as N_2O (*frac*),

~~2.2.3~~2.2.4 Responses to elevated CO_2 and temperature

~~The respones~~Respones of N_2O emissions to atmospheric CO_2 and global warming have been ~~intensively studied~~reported at field scale~~.~~ (Dijkstra et al., 2012;van Groenigen et al., 2011). Here, we evaluate the model's response to step changes in form of a doubling of preindustrial CO_2 level (284 ppm to 568 ppm) and a 2K increase in atmospheric temperature. Starting from the same quasi-~~equalibrium~~equilibrium state with potential vegetation ~~obtained~~as in the global hindcast experiment in 2.2.1, we conducted four transient model runs: (1) the CONTROL run with the same drivers as spin-up; (2) the CO2_FERT run with the same drivers as the CONTROL except a doubling of atmospheric CO_2 level; (3) the TEMP run with the same drivers as the CONTROL except a 2K rise in atmospheric temperature; and (4) the CO2_FERT×TEMP run with both the doubling of CO_2 and 2K rise in temperature. For each experiment, we ran the model for 100 years and evaluated the corresponding results.

2.3 Comparisons with observations ~~and correlations with environmental variables~~

We compared our model results for annual N_2O gas loss with field data: We compiled annual N_2O emissions from peer-reviewed literature (see Appendix ~~B~~A for more information). To increase the representativeness of the measurements, we included only sites with more than 3 months or 100 days experimental span. We limited our datasets where there was no reference to a disturbance of any kind. Only locations with at least 50 years non-disturbance history for forests and 10 years for vegetation other than forests were included. The compiled 61 measurements cover a variety of spatial ranges with ~~the~~ vegetation ~~type~~types including tropical rainforest, temperate forest, boreal forest, tundra, savanna, perennial grass, steppe, alpine grass and desert vegetation. Multiple measurements ~~from~~falling into the same ~~corresponding~~model grid cell ~~in the model~~ were averaged. If the authors had indicated the dominant vegetation or soil type, we used the values reported for the dominant type instead of the averaged. For multiyear measurements ~~from the same study~~, even if the authors gave the individual year's

data, we averaged the data to avoid overweighting of long term studies. If the location was between borders of different model grid cells, we averaged across the neighboring grid cells.

~~Pearson correlation coefficient with the significance threshold of $\alpha <$~~ We also compared monthly N_2O fluxes at a group of sites: (a) the Tapajós National Forest in Amazonia (3°S, 55°W), taken from Davidson et al. (2008); (b) the Hubbard Brook Experimental Forest in New Hampshire, USA (44°N, 72°W), as described in Groffman et al. (2006); (c) the cedar forest from Oita, Japan (33°N, 131°E), as described in Morishita et al. (2007); (d) the *Leymus chinensis* (LC) and *Stipa grandis* (SG) steppe in Inner Mongolia, China (44°N, 117°E), taken from Xu-Ri et al. (2003); (e) the cedar forest in Fukushima, Japan (37°N, 140°E), taken from Morishita et al. (2007); and (f) the primary (P1 and P2) and secondary (L1 and L2) forests located at the Pasir Mayang Research Site (1°S, 102°E), Indonsia, taken from Ishizuka et al. (2002). In addtion, daily measurements of soil temperature, soil moisture and N_2O emissions were compared at four German forest sites located in the same grid cell (50°N, 8°E), as described in Schmidt et al. (1986). ~~was used to quantify the correlation between N_2O fluxes and environmental variables, i.e. soil temperature, root zone water content, gross primary productivity, net mineralization rate, soil ammonium and soil nitrate content, for each grid cell from the global hindcast run.~~

3 Results

3.1 Global budget, seasonal and inter-annual variability

Our modelled global soil N_2O flux is 6.~~82~~69±0.~~28~~32 TgN yr^{-1} (1970-2005 mean and standard deviation among different years) (Fig.1) with LM3V-SM (Method 3, default method for LM3V-N calculated soil moisture), 5.61±0.32 TgN yr^{-1} with NOAH-SM (Method 2) and 7.47±0.30 TgN yr^{-1} with ERA-SM (1982-2005, Method 2) which is within the range of reported values: The central estimation of N_2O emission from soils under natural vegetation is 6.6 TgN yr^{-1} based on the Intergovernmental Panel on Climate Change (IPCC) AR5 (Ciais et al., 2013) (range, 3.3–9.0 TgN yr^{-1}) for the mid-1990s. Mean estimation for the period of 1975-2000 ranged from 7.4 to 10.6 TgN yr^{-1} with different precipitation forcing data (Saikawa et al., 2013). ~~Xu et al. (2012)~~Xu-Ri et al. (2012) reported the decadal-average to be 8.3-10.3 TgN yr^{-1} for the 20th century. ~~Potter and Klooster (1998)~~Potter and Klooster (1998) reported a global mean emission rate of 9.7 TgN yr^{-1} over 1983-1988, which is higher than the earlier version of their

model (6.1 TgN yr^{-1}) ~~(Potter et al., 1996)~~(Potter et al., 1996). Other estimates includes 6-7 TgN yr^{-1} (Syakila and Kroeze, 2011), 6.8 TgN yr^{-1} based on the O-CN model (Zaehle et al., 2011), 3.9-6.5 TgN yr^{-1} for preindustrial periods from a top-down inversion study ~~(Hirsch et al., 2006)~~(Hirsch et al., 2006), 1.96-4.56 TgN yr^{-1} in 2000 extrapolated from field measurements by an artificial neural network approach (Zhuang et al., 2012), 6.6-7.0 TgN yr^{-1} for 1990 (Bouwman et al., 1995), and 7-16 TgN yr^{-1} (Bowden, 1986) as well as 3-25 TgN yr^{-1} (Banin, 1986) from two earlier studies.

Following Thompson et al. (2014), El Niño years are set to the years with the annual multivariate ENSO index (MEI) greater than 0.6. 1972, 1977, 1982, 1983, 1987, 1991, 1992, 1993, 1994, 1997 and 1998 were chosen as El Niño years. We detected reduced emissions during El Niño years (Fig. 1), in line with the global atmospheric inversion study of Thompson et al. (2014) and the process based modelling study from ~~Saikawa et al. (2013)~~Saikawa et al. (2013).

Figure 2 shows the simulated global natural soil N_2O emissions in 4 seasons averaged over the period of 1970-2005~~.~~ based on LM3V-SM (Method 3). The northern hemisphere displays a large seasonal variability, with the highest emissions in the northern summer (JJA, June to August) and lowest in winter (DJF, December to February). Globally, northern spring (MAM, March to May) has the highest emission rate (2.07 TgN) followed by summer (1.89 TgN). The smaller emissions in summer compared to spring stems from a reduced contribution of the southern hemisphere during northern summer.

As expected, a large portion (more than 60%) of the soil N_2O fluxes have tropical origin (23.5 S to 23.5N), while emissions from cooler regions are limited by temperature and arid/semi-arid regions by soil water. Our modelling results suggested year-round high emission rates from humid zones of Amazonia, east central Africa, and throughout the islands of Southeast Asia, with small seasonal variations (Fig. 2). Emissions from tropical savannah are highly variable, with locations of both high fluxes (seasonal mean > ~~90~~30 mgN m^{-2} ~~season~~month^{-1} or 3.6 kg ha^{-1} yr^{-1}) and low fluxes (seasonal mean < ~~4~~1.3 mgN m^{-2} ~~season~~month^{-1} or 0.16 kg ha^{-1} yr^{-1}). The simulated average tropical emission rate is 0.78 kgN ha^{-1} yr^{-1} (1970-2005), within the range of estimates (0.2-1.4 kgN ha^{-1} yr^{-1}) based on site-level observations from the database of ~~Stehfest and Bouwman (2006)~~Stehfest and Bouwman (2006), but smaller than a more detailed simulation study (1.2 kgN ha^{-1} yr^{-1}) carried out by ~~Werner et al. (2007)~~Werner et al. (2007).

Our analysis here excluded land cover, land use changes and human management impacts, while most of the observation-based or regional modelling studies did not factor out those impacts. Our modelling result in natural tropics is comparable with another global modelling study (average emission rate, 0.7 kgN ha^{-1} yr^{-1}) ~~(Zaehle et al., 2010)~~(Zaehle et al., 2010), in which the authors claimed they ~~might~~may underestimate the tropical N_2O sources compared to the inversion estimates from the atmospheric transport model TM3 (Hirsch et al., 2006).

3.2 Sensitivity to WFPS

~~Soil~~The different parameterization of WFPS and the use of different soil moisture modeling and data allows to test the sensitivity of soil N_2O emissions to variable WFPS. Globally, emissions generally increase with WFPS (Fig. ~~4~~3). WFPS derived from Method 1 is higher than that based on Method 2. ~~Soil~~Data-derived soil moisture datasets ~~and~~combined with different calculation methods together produced a range of 0.15-0.72 for the global mean WFPS (1982-2005). ~~Mean value~~ While mean values greater than 0.6 (approximately field capacity) are less realistic, ~~though~~ these high WFPS values provide ~~us~~ the ~~oportunity~~opportunity to test the model's ~~sensitivity~~response to the soil moisture-based parameterization of redox conditions in soils. Global soil N_2O emissions are highly sensitive to WFPS, with approximately 0.25 TgN per year per 0.01 change in global mean WFPS in the range 0 to 0.6. The spatial and temporal characteristic of WFPS also matters. ~~With mean WFPS of *ca.* 0.21, emission~~Emission rate from LM3V-SM (Fig. ~~4~~3 green cycle) is 1.13 TgN yr^{-1} higher than that from NOAH-SM (Fig. ~~4~~3 blue triangle), ~~showing~~while both model configuration have the same mean WFPS (ca 0.21), highlighting effects of regional and temporal differences between the soil moisture products.

3.3 Model-observation comparisons

Modelled N_2O emissions capture the average of cross-site ~~observation~~ observed annual mean emissions (0.54 vs. 0.53 kgN ha^{-1} yr^{-1} based on LM3V-SM ~~with WFPS Method 3~~) reasonably ~~at the annual time step~~ (Appendix ~~B~~A and Fig. ~~3a~~4a), but spread considerably along the 1:1 line. The points deviating the most are from tropical forests, with overestimations from montane tropical forest and underestimations from lowland tropical forests if those measurements are representative of gridcell emissions. These patterns are similar as results from NOAH-SM (Appendix ~~B~~A and Fig. ~~3b~~4b) and ERA-SM (Appendix ~~B~~A and Fig. ~~3c) with WFPS based on Method 2,~~4c), except that the application of WFPS from NOAH-SM slightly underestimates

the observed global mean (0.54 vs. 0.47 kgN ha^{-1} yr^{-1} from NOAH-SM with WFPS based on Method 2).

~~3.4 Correlations with environmental variables~~

~~Figure 5 illustrates the temporal correlations between simulated monthly soil N_2O emissions and environmental variables (surface soil temperature, root zone soil water content, gross primary productivity, net mineralization rate, soil ammonium content and soil nitrate content), which were either predicted by the model or model inputs (forcings). The results show, that temperature is a strong driver of N_2O emissions in boreal and across large swaths of temperate regions. Temperature directly affects nitrification and denitrification rates, and also alters the N made available from mineralization and competition with plant uptake. Higher temperature triggers N_2O emissions from boreal and a large fraction of temperate ecosystems, while both positive and negative temperature relationships exist in tropical forests (Fig. 5a). Covariation with soil temperature result in a strong postive link between gross primary productivity, net mineralization rate and N_2O emission in the northern high latitudes (Fig. 5d). Likewise, higher root zone water content is associated with higher soil N_2O emissions except in the northern mid- to high latitudes where soil temperature is the primary controller (Fig. 5a,b). Tropical forests and some of the humid temperate regions with high N_2O emissions show the strongest soil moisture-N_2O flux correlations, which partly explains the high sensitivity of global soil N_2O budget to WFPS.~~

~~As expected, N_2O emissions are strongly and positively correlated with soil nitrate content at the global scale (Fig. 5f), while the relationships between N_2O emissions and soil ammonium (Fig. 5e) varies. In the humid tropics, N_2O fluxes are negatively correlated with soil ammonium content. This negative pattern results from the inverse relationship between soil ammnium content and nitrification rate. In our model, soil ammonium content is not only constrained by temperature or moisture but is also subjected to varying biological demand from plants and microbes. For example, high specific nitrification can draw down ammonium concentration in the soil.. Compared to the humid tropics, soil ammonium levels in cold or dry areas appear to be mainly controlled by N supply (mostly from SOM decomposition/N mineralization). In cold (or dry) regions, SOM decomposition/N mineralization, nitrification and denitrification are all regulated by soil temperature (or moisture) (Fig. 5a). The correlation between soil ammonium and N_2O fluxes covaries with the soil temperature (or moisture)-N_2O flux relationship.~~

At the Tapajós National Forest, results from LM3V-SM capture some of the variations in N_2O fluxes, but the model is not able to reproduce the high emissions observed during spring (Panel (a), Fig. 5). At the Hubbard Brook Experimental Forest, the correlation between model results and observations are 0.51 (LM3V-SM), 0.56 (NOAH-SM) and 0.62 (ERA-SM) for yellow birch, 0.66 (LM3V-SM), 0.68 (NOAH-SM) and 0.70 (ERA-SM) for sugar maple, However, the model is less robust in reproducing the magnitude of emission peaks. Groffman et al. (2006) suggested high emissions of N_2O in winter were associated with soil freezing. However, the model assumes little emissions when soil temperature is under 0 °C. In addition, observations suggested N_2O uptake (negative values in Panel (b), Fig. 5) while the model does not incorporate mechanisms to represent N_2O uptake. At the Oita cedar forest, model reproduces the seasonality of N_2O emissions accurately (Panel (c), Fig. 5). ERA-SM overestimates the magnitude of N_2O fluxes from Inner Mongolia grassland, while the magnitudes produced from LM3V-SM and NOAH-SM are comparable with observations. However, the timing of the emission peaks are one or two month in advance from model output compared to observations (Panel (d), Fig. 5). At the Fukushima cedar forest, similar as at the Oita cedar forest, models are less robust at capturing the magnitude of high peaks despite the seasonality produced by the model are good (Panel (e), Fig. 5). Emissions from the primary and secondary tropical rainforest at the Pasir Mayang Research Site are highly variable, which makes the comparison difficult (Panel (f), Fig. 5). LM3V-SM (but not ERA-SM and NOAH-SM) reproduces the low emissions in September-November 1997 and the increase of emissions from secondary forests in December, 1997. Overall, modeled variability is smaller compared to observation.

The strong variability of measured N_2O emissions is further illustrated in Fig. 6. Difference in measured N_2O fluxes between different forest sites within one grid cell is large, reflecting the heterogeneity that is not captured within one grid cell. In addition, the error bars, which represent the standard deviation of measured N_2O fluxes at three different plots of the same forest, are large. The standard deviation is as high as 49.27 $\mu gN\ m^{-2}h^{-1}$, indicating the strong variability of measured N_2O fluxes at the plot scale. Modeled N_2O fluxes are generally within the range of measured N_2O emissions. Model outputs slightly underestimate N_2O emissions largely due to the underestimation of soil water content (Panel (b) Fig. 6).

3.4 Sensitivity to N cycling processes and parameterization

Disallowing of N losses through DON and fire volatilization enhance ecosystem N accumulation and availability to plants and microbes, and therefore increases N_2O emissions (Panel (a), Fig.7). The gain in N_2O emissions from disallowing DON loss is small (0.12 TgN yr^{-1}). However, N_2O emission is on average (1950-2005) increased by 3.63 TgN yr^{-1} in the absence of fire volatilization N loss (we note, that fires do occur, but N is retained as ash in the litter). The gain is most evident in tropical regions (not shown), indicating the importance of fire in regulating ecosystem N status. Simulated preindustrial BNF is smaller than the empirical reconstructed BNF (72 in LM3V-N vs. 108 TgN yr^{-1} from empirical based data). However, BNF in LM3V-N increases with time under historical varying climate, increasing atmospheric CO_2 level and N deposition. The global average BNF during 1950-2005 is 100 TgN yr^{-1}, close to the empirical value. Neverthless, substitution of BNF in LM3V-N by empirical preindustrial value increased N_2O flux by 1.2 TgN yr^{-1}(Panel (a), Fig.7).

Among the specific parameters tested, N_2O emission is most sensitive to the 10 times change (x10) of the fraction of net nitrification lost as N_2O gas. The relative magnitude of N_2O flux on average (1950-2005) reaches 6.5 times of the default (Panel (b), Fig.7). Reduction (x0.1) of maximum active plant N uptake strength (v_{max}) strongly increases N_2O emissions (*ca.* by 3 times of the default). Meanwhile, enhancement of v_{max} also increases N_2O fluxes, reflecting the non-linear response of N_2O emissions to v_{max}. x10 in the maximum nitrification rate k_n and denitrification rate k_d increase N_2O emissions, while x0.1 decrease N_2O flux. N_2O increases more with increasing k_d than with increasing k_n, whereas reduction of k_n (x0.1) produces a stronger response than reduction of k_d. The half-saturation constant that represents the regulation of labile carbon availability on denitrification rate, *Kc*, is the least sensitive parameter. Meanwhile, reduction (x0.1) of the half-saturation constant *Kn* that represents the regulation of substrate availability on denitrification rate on average increased N_2O fluxes by 4.5 TgN yr^{-1}(Panel (b), Fig.7).

3.5 CO_2 and temperature responses

Globally, N_2O emissions respond to a step CO_2 increase first with a decline to ultimately increased levels after approximately ~~45~~40 years (Fig. ~~6a~~8a, black line). The simulated global response follows largely the behaviour as simulated for tropical forests (Fig. ~~6a~~8a, yellow line). The shift from a negative to a positive response indicates possible competing mechanisms

operating on different time scales. Field level experiments revealed the highly variable effects of CO_2 fertilization on N_2O emissions. ~~From~~Based on a meta-analysis, van Groenigen et al. (2011) suggested that elevated CO_2 significantly increased N_2O emission by 18.8%, while ~~Dijkstra et al. (2012)~~Dijkstra et al. (2012) argued for a non-significant response in non-N-fertilized studies. In contrast to observation studies, ~~two~~the global C-N cycle model analyses from O-CN suggested negative ~~effects from~~ CO_2 fertilization effects on N_2O emissions ~~(Xu et al., 2012;Zaehle et al., 2011)~~(Zaehle et al., 2011). The negative impacts (reduced N_2O flux), which are also reported ~~from~~in manipulative experiments, are likely from increased plant N and immobilization demand under CO_2 fertilization, reducing N availability for nitrifiers and denitrifiers~~. Positive~~ (Dijkstra et al., 2012). CO_2 fertilization on average (over 100 years) increased the global mean plant nitrogen uptake rate by 10.02 kgN ha^{-1} yr^{-1}, as shown in Fig. 9 (Panel (b)). Modelled soil inorganic N content (ammonium and nitrate) is reduced at first, but the reduction is not sustained. One mechanism to alleviate CO_2 fertilization caused N limitation is through BNF, which is on average (over 100 years) more than doubled (Fig. 9 Panel (e)). Similar as manipulative field experiments (Dijkstra et al., 2012), positive effects (increase N_2O fluxes) can result from the impacts of elevated CO_2 level to increase litter production (Fig. 9 Panel (a)) and consequently C sources for denitrifiers, and to increase soil moisture (Fig. 9 Panel (d)) from reduced stomatal conductance and leaf transpiration~~.~~ (Fig. 9 Panel (c)).With both ~~of these~~ positive and negative mechanisms embedded in our model, the net effects depend on the relative strength of ~~those~~the opposing forces.

Temperate deciduous forests, where most of the forest CO_2 fertilization experiments are conducted, respond positively to elevated CO_2 level (Fig. ~~6a~~8a, green line). The slight increase in modelled N_2O emission are comparable with the mean response of field data compiled for temperate forests (*ca.* 0.01-0.03 kgN yr^{-1} ha^{-1}) ~~(Dijkstra et al., 2012)~~(Dijkstra et al., 2012). A similar positive response was detected for cold evergreen forests (Fig. ~~6a~~8a, pink line) with stronger magnitude compared to temperate deciduous forests. For grasslands, ~~Dijkstra et al. (2012)~~Dijkstra et al. (2012) reported small negative mean response from northern mixed prairie (ΔN_2O, *ca.* -0.01 to -0.03 kgN yr^{-1} ha^{-1}), zero mean response from shortgrass steppe and positive mean response from annual grassland (*ca.* 0.03-0.06 kgN yr^{-1} ha^{-1}). Our model shows a small negative mean response from C4 grassland (Fig. ~~6a~~8a, cyan line) with the similar magnitude of that reported for the Northern mixed prairie, where the composition of C4 grass varies ~~(Dijkstra et al., 2012). A CO_2 increase in C3 grassland initially reduces N_2O emission (Fig. 6a, blue line).~~

~~However, this slight negative response turns into a small positive after one or two decades.~~(Dijkstra et al., 2012). A CO_2 increase in C3 grassland initially reduces N_2O emission (Fig. 8a, blue line). However, this slight negative response turns into a small positive within one decade.

Elevated temperature generally increases N_2O emissions except for the slight negative effect in C4 grass (Fig. ~~6b~~8b). Overall the response to a 2 degree warming is bigger than that of doubling of CO_2. The simulated temperature effects are more pronounced in the first decade and decrease over time in tropical forests (Fig. ~~6b~~8b, yellow line), while for the temperate deciduous forests (Fig. ~~6b~~8b, green line) and boreal forests (Fig.~~6b~~8b pink line), the temperature effects become more pronounced over time. Simulated temperate forest response (in the first decade) is close to that of observed mean (*ca.* 0.2-0.5 kgN yr^{-1} ha^{-1}) ~~(Dijkstra et al., 2012).~~(Dijkstra et al., 2012). Our modelled slight negative response in C4 grass and positive in C3 grass are in alignment with data compiled by Dijkstra et al. (2012) who reported both positive and negative responses in grasslands.

The results of combining CO_2 and temperature are similar to the CO_2 effect alone (Fig. ~~6c~~8c), despite the fact, that the individual effect of temperature is much stronger than that of CO_2. This antagonistic interaction (i.e. the combined enhancement in N_2O flux from elevated CO_2 and temperature are smaller than the summary of their individual effects) is also evident for C3 grass (first 50 years), temperate deciduous tree and cold evergreen forests (Fig. ~~6d~~8d).

4 Discussion

Our model combines ~~knowledge from~~ two of the most widely applied biogeochemical models (DNDC and CENTURY) with current advancements in field level studies. ~~Our global~~The model is capable of reproducing the global mean natural N_2O emissions from other modeling and inverse methods, and the average of observed cross-site annual mean behavior. By focusing on the role of soil moisture in N_2O emissions, we find a global scale ~~(not surprisingly)~~ high dependence of simulated N_2O emissions on soil moisture (WFPS), mainly driven by emissions from tropical regions. The model broadly reproduces the magnitude and direction of responses to elevated CO_2 and temperature from manipulative field experiments where data ~~are~~is avilable. The global responses to elevated CO_2 and temperature follow largely the response of tropical forests, where a noted absence of field experiments ~~exsit. Next, we will further discuss modelled responses to soil moisture and elevated atmospheric CO_2 and temperature.~~exist.

Evaluation of global simulations agaist field measurements is susceptible to scale mismatches. The complexity of microscale interactions for N_2O production creates notorious large spatial and temporal variabilities which are undoubtedly difficult to constraint even at the stand level (Butterbach-Bahl et al., 2013). ~~The~~Daily measurements from the German forest sites (Fig.6) illustrate the large variability in N_2O emissions. Further improvement in soil moisture simulation will improve our estimation of N_2O fluxes at the German forest sites. However, the homogeneous representation of environmental drivers within model grid cells casts doubt on site-specific model-observation comparison in global simulations. For example, N_2O emissions vary with topography which are not treated explicitly in most of the global C-N models. 3.8 times difference was detected in a montane forest (Central Sulawesi, Indonesia) moving from 1190 m to 1800m ~~(Purbopuspito et al., 2006)~~(Purbopuspito et al., 2006), and 4.3 times difference was found from a tropical moist forest (Brazilian Atlantic Forest) with the altitude change from 100m to 1000m ~~(Sousa Neto et al., 2011)~~(Sousa Neto et al., 2011)~~. Nevertheless, modeling approaches can offer important insights with respect to scaling our understanding of the mechanism of N_2O gas emissions to the globe.~~. However, comparison against field data revealed, that the model's varibility is smaller compared to observation for both across field sites (Fig. 4), and at different sites (Figs. 5 and 6). One of the reason for this shortcoming may be that fast transitions, such as freeze-thaw cycle (Groffman et al., 2006) and pulsing (Yienger and Levy, 1995) are not sufficiently captured.

Soil moisture is a key variable in climate system but difficult to derive or measure at the global scale ~~(Seneviratne et al., 2010)~~(Seneviratne et al., 2010). Our modelled fluxes are highly sensitive to WFPS, which is in agreement with observation and model synthesis studies ~~(Heinen, 2006~~(Heinen, 2006;Butterbach-Bahl et al., 2013). The large range when calculating WFPS from different methods resulted in a difference of more than 5 TgN yr^{-1} in global soil N_2O fluxes. ~~Saikawa et al. (2013)~~Saikawa et al. (2013) found an up to 3.5 TgN yr^{-1} gap induced by different precipitation forcing data from CLMCN-N2O. It is difficult to single out the difference caused by soil moisture alone from their results. Nevertherless, those two studies did suggest the importance of improving the dynamics of soil water and representation of WFPS for the purpose of predicting soil N_2O emission and climate feedbacks.

The root zone soil water in LM3V-N is based on a single layer bucket model. This simplified treatment of soil water dynamics may increase the difficulty in reproducing the temporal and spatial dynamics of WFPS. As a first step, we used the average between the ~~oringinal~~original

analog in LM3V-N and ~~that~~thatis derived from soil total porosity to account for actual soil moisture and the possibility of soil water above field capacity. Meanwhile, ~~with~~overriding soil moisture ~~replace treatments~~with data-derived products (NOAH-SM and ERA-SM), ~~WFPS based on method 2 (total porosity) is more close to real WFPS, indicating~~suggests that the most realistic average (1970-2005) soil N_2O emission is in the range of 5.~~74~~61-7.47 TgN yr^{-1}. A more realistic root zone water module, such as multilayer representations of biogeochemistry and soil water dynamics, would refine models of soil N_2O emissions. El Niño events trigger reduced soil emissions in our results similar as proposed by ~~Saikawa et al. (2013)~~Saikawa et al. (2013) and Thompson et al. (2014). El Niño events are known to have induced several of the most well known large scale droughts and alters soil moisture dynamics ~~(Schwalm et al., 2011)~~(Schwalm et al., 2011). Tropical forests N_2O emissions are highly correlated with root zone soil water content and contribute strongly to the global-scale fluxes of N_2O in our model. Whether there is a strong link between soil N_2O emission anomalies and El Niño induced soil moisture deviations needs further investigation with improved soil hydrology.

~~Patterns of seasonality, and the correlates between N_2O emissions vs. temperature and soil moisture suggest that moisture is the dominant driver of N_2O emission in tropical regions and soil temperature critical elsewhere. However, globally~~Globally, the tropical fluxes contribute with more than 60% to the global soil N_2O fluxes. Also, global responses to elevated CO_2 and temperature are dominated by the tropical response. In contrast to temperate and boreal forests, tropical forests respond negatively to elevated CO_2 in the first few decades. Our results therefore suggest caution when extrapolating from current manipulative field studies to the globe: The postive response to CO_2 enrichment as obtained from (mostly) extratropical field study may be overestimated, when the studies' fluxes are scaled up to the globe. Moreover, we found strong interaction of elevated CO_2 and temperature, acting to reduce soil N_2O emission compared to the sum of individual responses, highlighting the non-linear impacts of CO_2 and temperature on N_2O emissions. ~~We realize that this interacion~~Our results from step increases of CO_2 and temperature is different from Xu-Ri et al. (2012) in which CO_2 and climate change act synergistically to increase historical N_2O emissions, especially in tropical regions. CO_2 fertilization plus interaction with temperature rise reduce tropical N_2O fluxes in the first several decades from our model. We realize that this interaction is likely to be different when incorporating other factors ~~(Brown et al., 2012), such as N depostion~~(Brown et al., 2012), such as N depostion, precipitation and land use change (disturbance). In addition, step changes in

atmospheric CO_2 and temperature compared to gradual and sustained increases may also lead to differences, and may explain the discrepancy ~~to two of~~between the ~~global~~previous modeling ~~studies that suggested an overall positive response of soil N_2O emission to the effects of elevated~~ study and meta-analysis of manipulative field experiments with regard to CO_2 ~~and climate change~~fertilization responses ~~(Xu et al., 2012;Zaehle et al., 2011)~~(Zaehle et al., 2011; van Groenigen et al., 2011). However, step changes mimic most closely ~~–~~manipulative experiments. Nevertheless, the largest uncertainties lie in the tropical region where our model indicated strongest responses and strongest nonlinear interactions of elevated CO_2 and temperature.

Globally, N_2O emissions from nitrification-denitrification are similar to O-CN and LPJ-DyN as they are all derived from DNDC. Embedding an established N_2O emission module into LM3V-N enables evaluation of the response of N_2O emissions under different assumptions across models with respect to the dynamics of the larger plant-soil N cycle. Generally higher inputs from BNF or constraints on losses through organic N (fire, DON) enhance N_2O emissions. The representation of of BNF in models requires impromvent but we show here that different implementations are globally important for N_2O emissions. Similar, the magnitude of N lost through fire impacts N_2O emissions in fire prone regions, while N emission factors are poorly constrained globally (Andreae and Merlet, 2001). The strength of plant uptake of N poses a strong constraint on the avaiability of N for nitrification-denitrification losses as it can draw down N substantially (Gerber and Brookshire, 2014). A reduction of plant uptake strength allows for relatively more N allocated for denitrification. More surprising was the positive effect of a stronger plant uptake capacity on N_2O emissions: Enhanced plant uptake allow increased vegetation production, and an throughput through litterfall and mineralization in the long run, which ultimately may allow higher N_2O losses in lieu of other export pathways. In addition to those N cycling processes N_2O emission is highly sensitive to the fraction of N lost as N_2O from net nitrification. The fraction of N_2O lost from net nitrification is uncertain. Goodroad and Keeney (1984) suggested a value of 0.1-0.2% , while Khalil et al. (2004) reported a range of 0.16%-1.48% depending on the O_2 concentration. We applied a global constant of 0.4% in our default simulation, bearing in mind the large uncertainies associated with this parameter. We also note that this value has significant impact on global N2O emissions.

The response to increases in temperature and CO_2 is a consequence of both the direct effect of temperature on nitrification and denitrification, and indirect effects via water and mineral N

availability. The inital negative response of N_2O emissions to CO_2 fertilization from tropical forests produced by LM3V-N stems largely from the increased demand and uptake of mineral N due to enhanced vegetation growth under elevated atmospheric CO_2 level. Despite soil N availability has been reported to decrease, unchanged or increase from manipulative CO_2 enrichment experiments across extrotropical ecosystems (Reich et al., 2006; Drake et al., 2011; Reich and Hobbie, 2013), no empirical evidence is available in tropical forests. LM3V-N produced, on average, a reduced soil mineral N concentration in tropical forests initially. Consequencely, less N is available for gaseous losses. If gross mineralization is used as an indicator of the rate of N flow in the "hole-in-the-pipe" concept and gaseous losses are propotional to mineralization, the inital negative response is unlikely to be detected. We found increased mineralization rate with increased litterfall under elevated CO_2, while N availability is reduced from LM3V-N. The mineralization based approach is likely to predict an inrease of losses regardless of N limitation. In LM3V-N, N availability recovers as N cycling processes adjust to CO_2 fertilization, especially from BNF, but also via higher transient retention of N from deposition.

In addition to the uncertainties mentioned above, we simplified N_2O sources and processes, ignoring other microbial metabolic pathways and ~~aboitic~~abiotic processes that produce or consume N_2O. The global magnitude of those ignored process remains largely unexplored. We do not incorporate explicit mechanisms for N_2O emissions from freeze-thaw cycle or poorly drained soils (e.g.wetlands), the uptake of organic N etc., which ~~might~~are be globally important, especially with future climate changes. Considering those uncertainties and gaps, more studies are in need in order to ~~unstand~~undstand the terretrial N_2O emissions.

5 Conclusions

We present estimates of terrestrial soil N_2O fluxes under natural vegetation (1970 to 2005) based on ~~a new~~existing N_2O emission ~~module~~formulations embedded into the global C-N cycle model LM3V-N. To determine the sensitivity of the modelling result to soil water (WFPS), we replaced the root zone soil water with two other derived datasets and altered the way in which WFPS is calculated. Our best estimate of modelled global soil N_2O flux is ~~6.82±0.28~~5.61-7.47 TgN yr^{-1} (1970-2005 mean and interannual variability), within the range of current understanding of soil N_2O emissions, but highly sensitive to WFPS~~.~~, general N cycling and parameterization of N_2O losses through nitrification and denitrification. Improvement of soil

hydrology is likely to significantly reduce the large uncertainties associated with soil N_2O emission estimates. Although the simulated mean responses are in agreement with manipulative field studies where effects of elevated CO_2 and temperature were investigated, we found that the global response was dominated by tropical forest, where our model suggest a different response than the field studies carried out in temperate ecosystems.

Appendix A: Soil N_2O emission module

Gaseous losses so far were not differentiated from hydrological leaching in LM3V-N. In this part, we provide details on the nitrification-denitrification module which explicitly simulates N gaseous losses from nitrification and denitrification, as well as other process modifications compared to the original LM3V-N.

A1 Nitrification-Denitrification

Transformation among inorganic N speicies (ammonium and nitrate) occurs mainly through two microbial pathways: nitrification and denitrification. Our simulation of N_2O losses during nitrification-denitrification generally follows the "hole-in-pipe" concept (Firestone and Davidson, 1989) with more detailed treatment of the N flux pipes and the leaky holes (gaseous losses) in the pipes.

Although ongoing debate exists in whether nitrification rates might be well described by bulk soil ammonium concentration or soil N turnover rate (Parton et al., 1996;Zaehle and Dalmonech, 2011), we adopt the donor controlled scheme (ammonium concentration). In additon to substrate, soil texture, soil water filled pore space (WFPS, the percentage of soil pore space filled with water), and soil temperature are all well known regulators of nitrification. As a first order approximation, nitrification rate (N) is simulated as a function of soil temperature, NH_4^+ availability and WFPS,

$$N = k_n f_n(T) f_n(WPFS) \frac{N_{NH_4^+}}{b_{N,NH_4^+}} \quad \text{(A1)}$$

where k_n is the ammonium turnover rate (11000 $year^{-1}$, the same as in LM3V-N). b_{N,NH_4^+} is the buffer parameter for NH_4^+(10 in LM3V-N); $f_n(T)$ is the temperature response function and $f_n(WFPS)$ is the soil water response function following Li et al. (2000), with a optimum temperature for nitrification at 35°C. The effect of WPFS on nitrification is texture dependent, with most of the reported optimum value around 0.6 (Parton et al., 1996;Linn and Doran, 1984). We adopted the WFPS response function from Parton et al. (1996) with medium soil texture.

$$f_n(T) = \left(\frac{60 - Tsoil}{25.78}\right)^{3.503} \times e^{\frac{3.503 \times (Tsoil - 34.22)}{25.78}} \quad \text{(A2)}$$

$$f_n(WFPS) = \left(\frac{WFPS - 1.27}{-0.67}\right)^{\frac{1.9028}{0.59988}} \times \left(\frac{WFPS - 0.0012}{0.59988}\right)^{2.84} \quad \text{(A3)}$$

where *Tsoil* is the soil temperature in degree Celsius. Denitrification is controlled by substrate NO_3^- (electron acceptor), labile C availability (electron donor), soil moisture and temperature. The responses of denitrification to substrate and labile C availability follow Michaelis-Menten kinetics. Labile C availability is estimated by soil heterotrophic respiration (*HR*). Following LPJ-DyN (Xu and Prentice, 2008), denitrification is assumed to have a Q_{10} value of 2 when the soil temperature is between 15 and 25 °C. Soil moisture response function is based on Parton et al. (1996). Soil pH is reported to be an important indicator of chemodenitrification which occurs only in acidic soils (pH<5) under conditions of high nitrite concentration. However, its role for N_2O production is not well studied (Li et al., 2000) and we do not model the chemodenitrification explicitly.

$$D = f_d(T) f_d(WFPS) \frac{HR}{HR+K_c} \frac{NO_3^-}{NO_3^- + K_n} \quad \text{(A4)}$$

$$\text{And } NO_3^- = \frac{N_{NO_3^-}}{b_{NO_3^-}} \quad \text{(A5)}$$

where *D* is the denitrification, *Kc*, *Kn* are Michaelis-Menten constants taken from Li et al. (2000) (0.017 and 0.083 kgN m^{-3} respectively); $b_{NO_3^-}$ is the buffer parameter for NO_3^- (1 in LM3V-N); $f_d(T)$ and $f_d(WFPS)$ are soil temperature and water reponse function for denitrification given by the following two equations

$$f_d(T) = e^{308.56 \times \left(\frac{1}{68.02} + \frac{1}{Tsoil + 46.02}\right)} \quad \text{(A6)}$$

$$f_d(WFPS) = \frac{1.56}{12.0^{\left(\frac{16.0}{12.0^{(2.01 \times WFPS)}}\right)}} \quad \text{(A7)}$$

A2 Gaseous partitions from nitrification-denitrification

N_2O loss from net nitrification is a constant fraction of 0.4%. NO_x emission from nitrification is based on the NO_x: N_2O ratio ($R_{NOx:N2O}$). $R_{NOx:N2O}$ varies with gas diffusivity (D/D_0) (Parton et al., 2001), which is estimated from air filled porosity (*AFPS*) (Davidson and Trumbore, 1995)

$$R_{NOx:N2O} = 15.2 + \frac{35.5 \times ATAN\left[0.68 \times \pi \times \left(10 \times \frac{D}{D_0} - 1.68\right)\right]}{\pi} \quad \text{(A8)}$$

$$\frac{D}{D_0} = 0.209 \times AFPS^{\frac{4}{3}} \quad \text{(A9)}$$

where ATAN stands for the trigonometric arctangent function; *AFPS* is the air-filled porosity (1-WFPS), and π is the mathematical constant, approximately 3.14159.

During denitrification, the gaseous ratio between N_2 and N_2O ($R_{N2:N2O}$) is calculated following Del Grosso et al. (2000), which combines the effects of substrate (NO_3^-) to electron donor (HR, the proxy for labile C) ratio and WFPS.

$$R_{N2:N2O} = Fr\left(\frac{NO_3^-}{HR}\right) \cdot Fr(WFPS) \quad \text{(A10)}$$

With

$$Fr\left(\frac{NO_3^-}{HR}\right) = \max\left(0.16 \times k, k \times e^{\left(-0.8 \times \frac{NO_3^-}{HR}\right)}\right) \quad \text{(A11)}$$

$$Fr(WFPS) = \max(0.1, 0.015 \times WFPS - 0.32)$$

where k is a texture dependent parameter (Table A1) estimated from Del Grosso et al. (2000).

Table A1 Texture dependent parameter k estimated from Del Grosso et al. (2000)

Soil Texture	Coarse	Medium	Fine	Coarse medium	Coarse/ fine	Medium/ fine	Coarse/ medium/ fine	Organic
k	2	10	22	6	12	16	11	2

A3 Other modified processes

We also added NH_3 volatilization into LM3V-N. NH_3 volatilization in soil results from the difference between the equilibrium NH_3 partial pressure in soil solution and that in the air. Dissolved NH_3 is regulated by ammonium concentration and pH. The net flux of NH_3 from soil to the atmosphere varies with soil NH_3, moisture, temperature, therefore

$$NH_3 = f(pH) f_{NH3}(T)(1 - WFPS) \frac{N_{NH_4^+}}{b_{N,NH_4^+}} \quad \text{(A12)}$$

where NH_3 is the net ammonia volatilization flux from each modelling step; $f(pH)$ is the pH factor and $f(T)$ is the temperature factor which are given by the following two equations

$$f(pH) = e^{2 \times (pH_{soil} - 10)} \quad \text{(A13)}$$

$$f_{NH3}(T) = \min\left(1, e^{308.56 \times \left(\frac{1}{71.02} - \frac{1}{Tsoil + 46.02}\right)}\right) \quad \text{(A14)}$$

where pH_{soil} is the soil pH which is prescribed instead of simulated dynamically. $f(pH)$ and $f(T)$ follow largely on the NH_3 volatilization scheme implemented in the dynamic global vegetation model LPJ-DyN (Xu and Prentice, 2008).

Appendix ~~B~~A: Observed annual N_2O fluxes data

Annual N_2O fluxes data were compiled from peer-reviewed literature. We applied simple selection criteria (see the main text) to reduce the mismatches between model outputs and field measurements, bearing in mind the gaps between complex field conditions and idealized model forcings. Latitutes (Lat) and longitudes (Lon) in Table ~~B1~~A1 are based on model grids.

Table ~~B1~~A1 Observed annual N_2O emission data for model comparison

No	Country	Lon	Lat	Location	Veg Type	N_2O kgN $ha^{-1}yr^{-1}$			
						OBS	LM3V-N	NOAH	ERA
1	Australia	133.1	-12.3	Douglas Daly region	Savanna	0.02	0.15	0.25	
2	Australia	148.1	-37.3	Moe	Temperate forest	0.11	0.58	0.74	0.72
3	Australia	151.9	-27.3	South-east Queensland	Tropical forest	0.52	0.01	0.03	
4	Austria	16.9	47.8	Klausenleopoldsdorf	Temperate forest	0.62	0.64	0.52	0.53
5	Austria	9.4	47.8	Achenkirch	Temperate forest	0.35	0.54	0.48	0.47
6	Austria	13.1	47.8	Innsbruck	Temperate forest	0.08	0.42	0.36	0.31
7	Austria	16.3	48.2	Schottenwald and Klausenleopoldsdorf	Temperate forest	0.76	0.61	0.54	0.53
8	Brazil	-61.9	-2.3	Manaus	Tropical rain forest	1.9	1.6	1.68	1.56
9	Brazil	-61.9	-2.3	Manaus	Tropical rain forest	1.930	1.71	1.74	1.55
10	Brazil	-54.4	-4.8	East-central Amazonia	Tropical rain forest	2.1	1.34	2.19	1.57
11	Brazil	-46.9	-2.3	Paragominas	Rainforest	2.430	1.22	1.19	1.11
12	Burkina Faso	-1.9	10.3	Ioba	Savanna	0.6	0.03	1.32	
13	Canada	-80.6	50.3	Ontario	Boreal forest	0.04	0.11	0.14	0.12
14	Canada	-106.9	52.8	Saskatchewan	Boreal forest	0.28	0.01	0.01	0.01
15	Canada	-103.1	52.8	Saskatchewan	Boreal forest	0.07	0.21	0.17	
16	Canada	-106.9	52.8	Saskatchewan	Boreal forest	0.09	0.01	0.01	
17	Canada	-73.1	45.3	Mont St. Hilaire	Temperate forest	0.42	0.54	0.46	
18	China	91.9	35.3	Tibet	Alpine grassland	0.07	0	0	0
19	China	125.6	40.3	Changbai mountain	Alpine tundra, temperate forest	0.56	0.73	0.64	0.45
20	China	114.4	42.8	Inner mongolia	Temperate forest	0.73	0.1	0.14	0.71
22	China	133.1	47.8	Sanjiang Experimental Station	Freshwater marshes	0.21	0.34	0.35	0.34
23	Denmark	13.1	55.3	Solo	Temperate forest	0.29	0.27	0.42	0.06

24	Denmark	13.1	55.3	Denmark	Temperate forest	0.52	0.28	0.37	0.05
25	Ecuador	-80.6	-4.8	Bombuscaro	Tropical forest	0.3	1.02	0	
26	Finland	24.4	60.3	Southern	Boreal forest	0.78	0.62	0.35	0.17
27	Germany	9.4	50.3	Average	Temperate forest	0.57	0.6	0.53	0.5
28	Germany	9.4	52.8	Kiel	Temperate forest	0.4	0.48	0.53	0.52
29	Germany	9.4	47.8	Southwest	Temperate forest	0.93	0.56	0.51	0.49
30	Germany	13.1	47.8	Höglwald	Temperate forest	0.41	0.47	0.4	0.39
31	Germany	9.4	52.8	Average	Temperate forest	0.66	0.44	0.5	0.5
32	Germany	9.4	52.8	Harz mountains	Mire	0.25	0.48	0.56	0.52
34	Indonesia	103.1	-2.3	Jambi	Lowland tropical rainforest	0.260	0.44		
35	Indonesia	121.9	-2.3	Central Sulawesi	Tropical seasonal rain forest	0.800	1.73	2.31	1.7
36	Indonesia	114.4	-2.3	Central Kalimantan	Tropical forest	2.51	2	2.45	1.73
37	Italy	9.4	45.3	P.Ticino BoscoNegri	Temperate forest	0.18	1.38	2.8	1.82
38	Malaysia	110.6	-2.3	Sarawak	Mixed peat swamp forest	0.7	0.66	0.65	0.57
39	New Zealand	170.6	-44.8	New Zealand	Temperate forest	0.01	1.24	2.84	1.24
40	Norway	9.4	60.3	Norway	Temperate forest	0.73	0.52	0.52	0.38
41	Panama	-80.6	7.8	Gigante Peninsula	Tropical forests	1.6	0.2	0.39	0.39
42	Sweden	13.1	57.8	Southwestern	Temperate forest	0.07	1.86	1.67	
43	Sweden	13.1	57.8	Asa experimental forest	Undrained bog	0.65	0.36	0.45	0.36
44	UK	-1.9	55.3	Northumberland	Grassland	0.3	0.4	0.5	0.41
45	USA	-73.1	42.8	Harvard forest	Mixed hardwood	0.04	0.56	0.54	0.48
46	USA	-73.1	40.3	New York	Temperate forest	0.9	0.4	0.49	0.41
47	USA	-80.6	25.3	Florida	Marsh	1	0.45	0	
48	USA	-73.1	42.8	New Hampshire	Temperate forest	0.070	0.64	2.15	
49	USA	-106.9	35.3	New mexico	Temperate forest	0.06	0.41	0.51	0.43
50	USA	-118.1	45.3	Washington	Temperate shrub-steppe	0.15	0.02	0.02	0.02
51	USA	-114.4	37.8	Mojave desert	Perennial grasses	0.11	0.02	0.02	0.02
52	USA	-106.9	40.3	Wyoming	Sagebrush steppe	0.21	0.01	0.02	0.03
53	USA	-73.1	45.3	Northeastern	Temperate forest	0.18	0.05	0.04	0.05
54	USA	-69.4	45.3	Northeastern	Temperate forest	0.03	0.53	0.46	0.44
55	USA	-103.1	40.3	Colorado	Temperate steppe	0.14	0.37	0.53	0.4
56	USA	-88.1	42.8	Wisconsin	Grass	0.040	0.03	0.05	0.05
57	USA	-114.4	37.8	Nevada	Mojave desert	0.11	0.45	0.45	

58	USA	-110.6	32.8	Arizona	Sonoran desert	0.4	0.04	0.04	0.05	
59	USA	-118.1	45.3	Ft. Collins, Colorado	Temperate grassland	0.12	0.01	0.03	0.03	
60	Venezuela	-61.9	10.3	Venezuela	Savana	0.73	0.06	0.07	0.07	
61	Zimbabwe	31.9	-17.3	Harare	Miombo woodland savanna	0.51	0.83	1.61	0.57	

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Acknowledgements

The Soil moisture data used in this study were acquired as part of the mission of NASA's Earth Science Division and archived and distributed by the Goddard Earth Sciences (GES) Data and Information Services Center (DISC)

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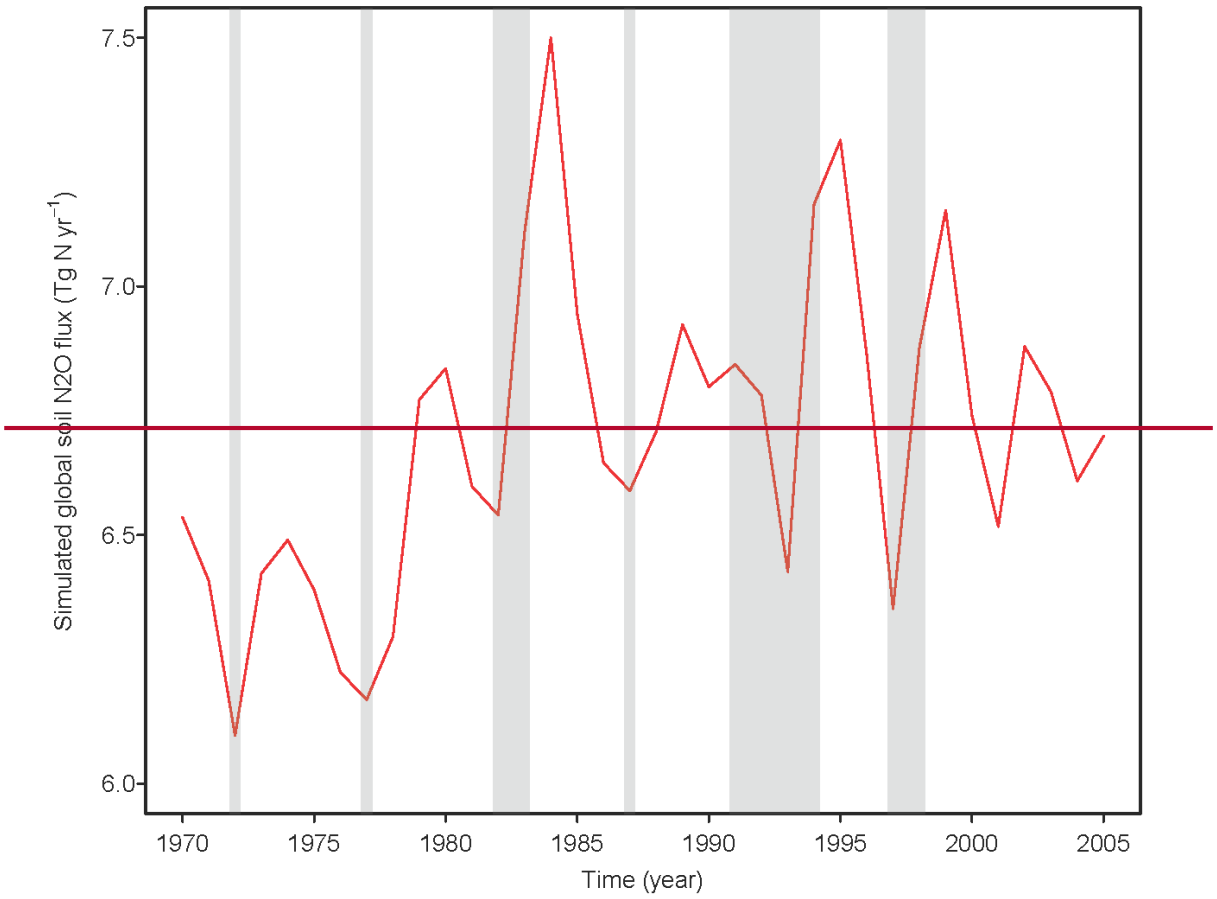

7.5
7.0
6.5
6.0
Simulated global soil N2O flux (Tg N yr^{-1})
1970
1975
1980
1985
1990
1995
2000
2005
Time (year)

1

2

3

4

Figures and Tables

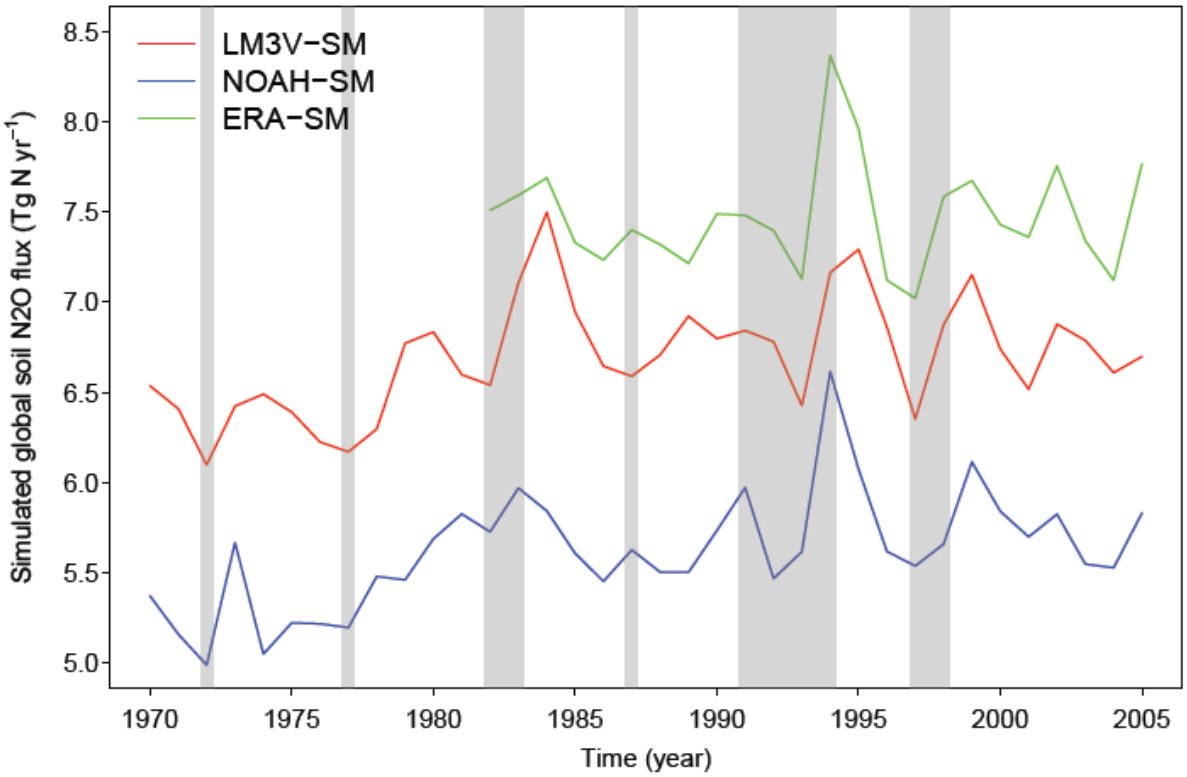


Figure 1. Simulated annual global soil N_2O emissions based on potential vegetation (1970-2005). Shaded grey area indicates El Niño years with the annual multivariate ENSO index (MEI) greater than 0.6. Colours refer to different soil moisture dataset used in the estimation: red for LM3V-SM (with WFPS calculated by Method 3); blue for NOAH-SM (Method 2) and green for ERA-SM (Method 2). Details for these soil moisture dataset and WFPS calculating methods is available in the main text.

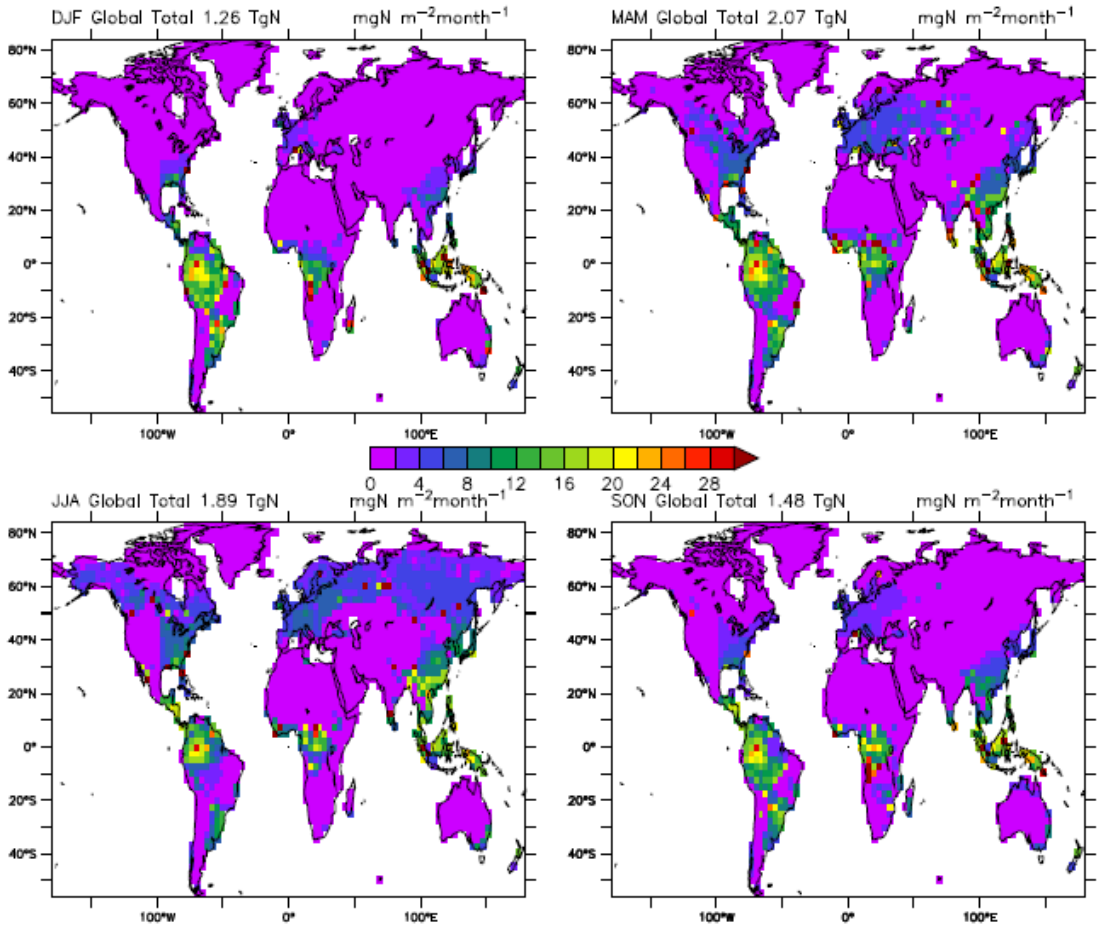


Figure 2. Global seasonal mean soil N_2O emissions (with potential vegetation) averaged over the years 1970-2005. DJF (December, January and February), stands for Northern Hemisphere Winter; MAM (March, April and May) for Spring; JJA (June, July and August) for Summer; and SON (September, October and November) for Autumn.

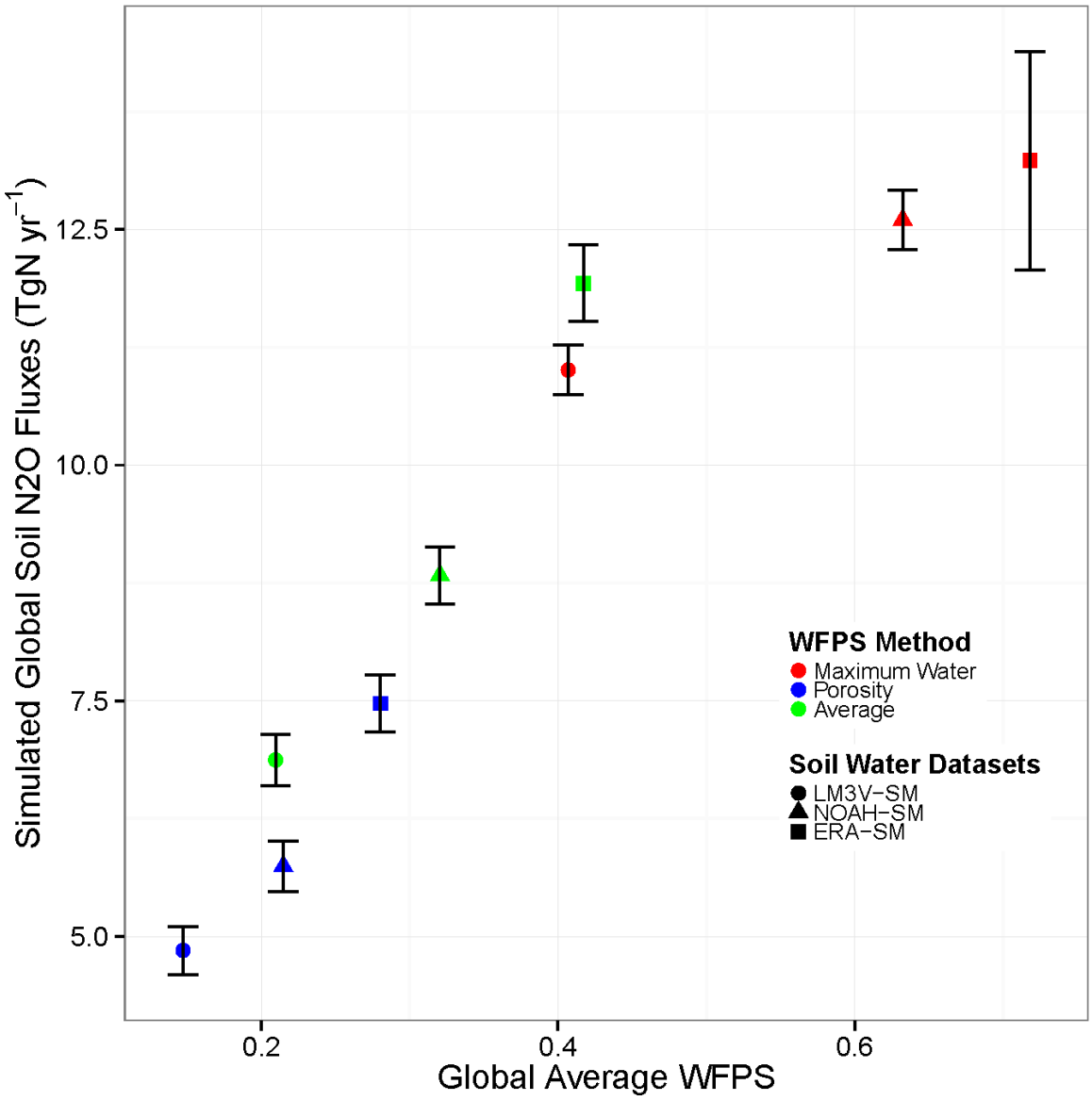


Figure 3. Sensitivity of simulated global soil N_2O emissions (with potential vegetation) to water filled pore space (WFPS). The x-axis is the WFPS averaged globally over 1982-2005; the y-axis represents the corresponding global total N_2O fluxes. A total of nine sets of WFPS are obtained through either different soil water datasets (colours) or varied calculation methods (symbols). Maximum water, porosity and average correspond to method 1, method 2 and method 3 in the main text, respectively. Coloured symbols represent interannual means and error bars indicate interannual standard deviations.

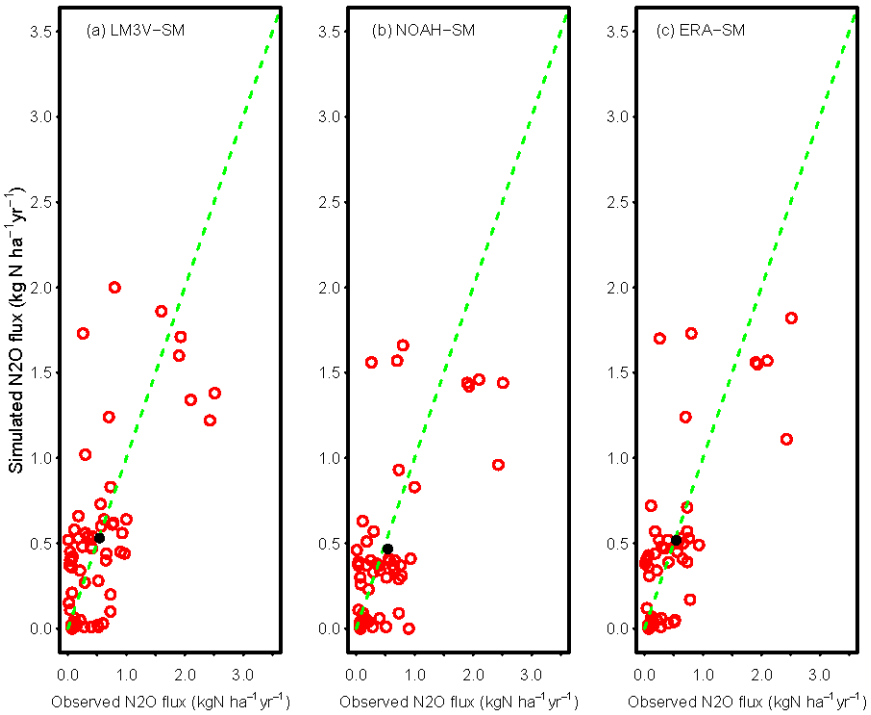


Figure 4. Observed vs. simulated annual N_2O emissions from natural soils. Dashed green lines are the 1:1 lines. The solid circles represent the overall means. Different panels represent simulations with different soil moisture data: (a) LM3V-SM (simulated by LM3V-N); (b) NOAH-SM (based on land surface model NOAH 3.3 in Global Land Data Assimilation System Version 2); and (c) ERA-SM (reanalysis data from ECMWF). Water filled pore space (WFPS) is calculated using the average of the one based on available water capacity and the one based on the total porosity (Method 3, see the main text for detailed description) for panel (a); and using the total porosity (Method 2) for panel (b) and (c).

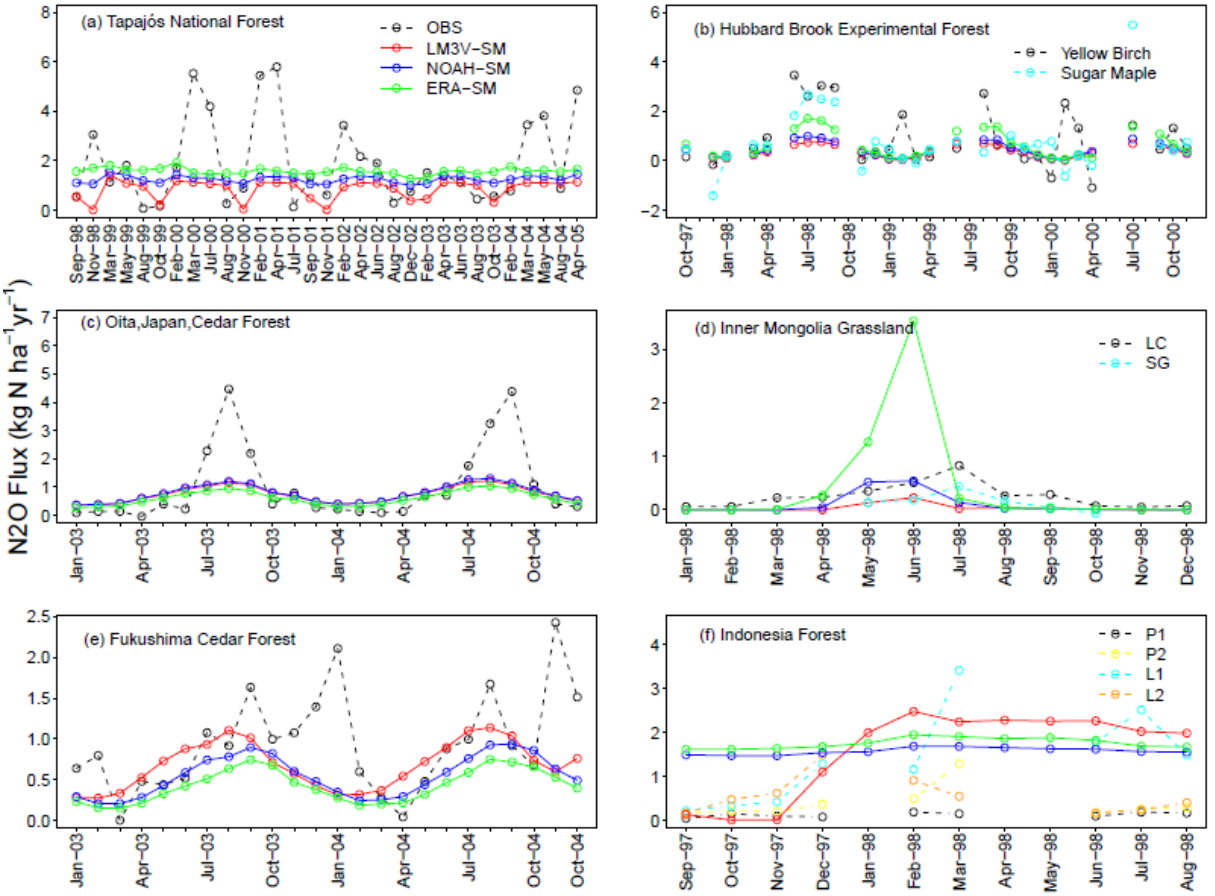


Figure 5. Observed vs. simulated monthly N_2O emissions at (a), the Tapajós National Forest in east-central Amazonia (3°S, 55°W), taken from Davidson et al. (2008); (b), the Hubbard Brook Experimental Forest in New Hampshire, USA (44°N, 72°W), taken from Groffman et al. (2006); (c), a cedar forest at Oita, Japan (33°N, 131°E), taken from Morishita et al. (2007) ; (d), the *Leymus chinensis* (LC)and *Stipa grandis*(SG) steppe in Inner Mongolia, China (44°N, 117°E), taken from Xu-Ri et al. (2003); (e), a cedar forest in Fukushima, Japan (37°N, 140°E), taken from Morishita et al. (2007); and (f), the primary (P1 and P2) and secondary (L1 and L2) forests located at the Pasir Mayang Research Site, Indonsia, taken from Ishizuka et al. (2002) (1°S, 102°E). Shown are modeled results from three WFPS schemes (LM3V-SM, NOAH-SM and ERA-SM) the same as in Figure 4.

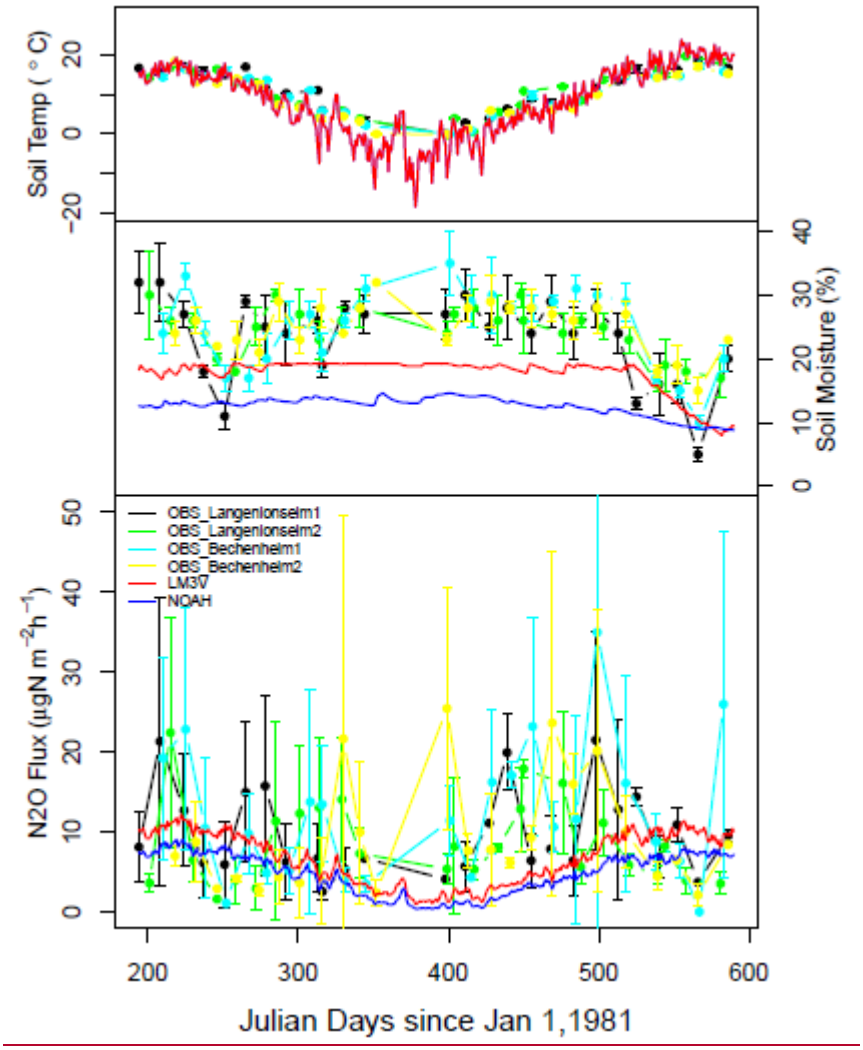


Figure 6. Comparison of (a) soil temperature (2cm from observation and 1 cm from model) in °C; (b) soil moisture (2cm from observation and root zone from model) in % and (c) soil N_2O emissions in µgN m^{-2} h^{-1} from observations and model outputs at four forest sites from German (50°N, 8°E), taken from Schmidt et al. (1986). Shown are modeled results from two WFPS schemes (LM3V-SM and NOAH-SM) similar as in Figure 4.

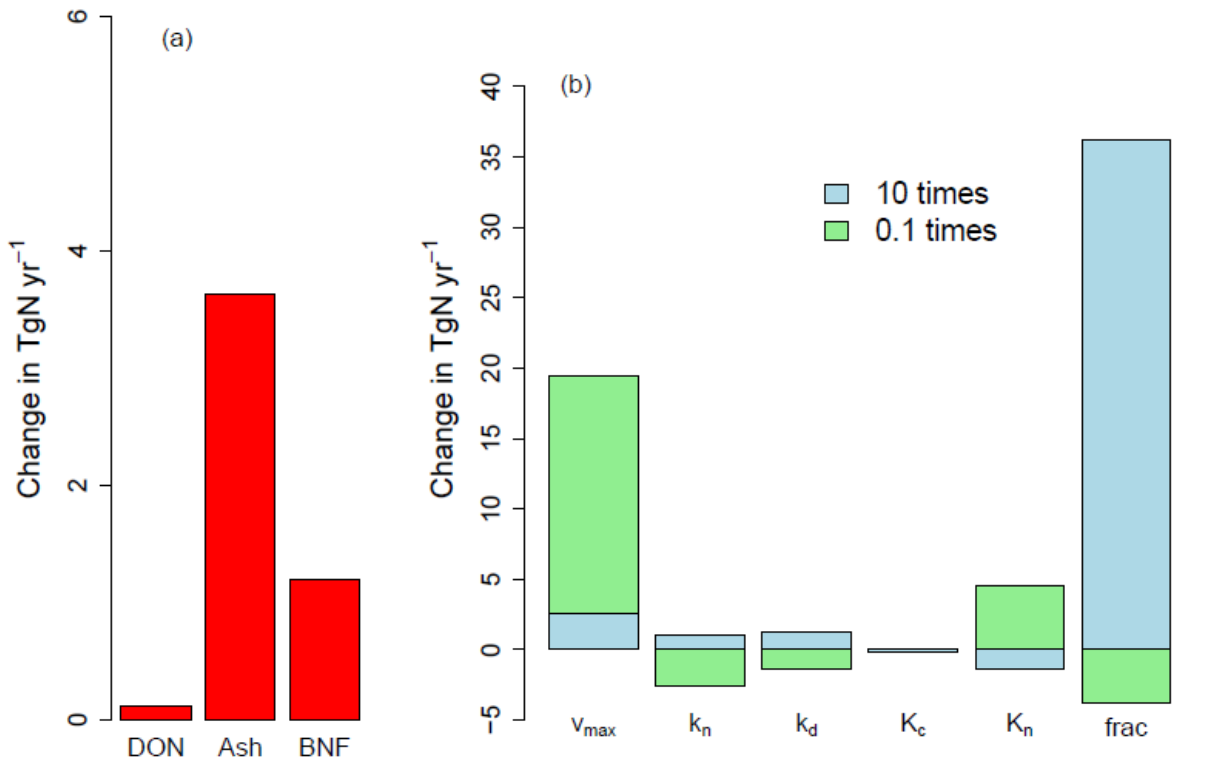


Figure 7. Changes in simulated global average N_2O (1950-2005) emissions from modifying general N cycling processes (a) and model parameters one-at-a-time (b). Altered processes include disallowing N losses through dissolved organic matter (DON in (a)) and fire volatilization (Ash in (a)), and replacing simulated biological N fixation with preindustrial N fixation rate (BNF in (a)). Parameters include: v_{max}, the maximum active N uptake rate per unit root biomass; k_n, the optimum nitrification rate; k_d, the optimum denitrification rate; *Kc* and *Kn* , the half saturation constants for labile C availability and nitrate respectively; and *frac* is the fraction of net nitrification lost as N_2O. Parameters are either increased by multiplying 10 (lightblue) or reduced by multiplying 0.1 (lightgreen) relative to the defaults .

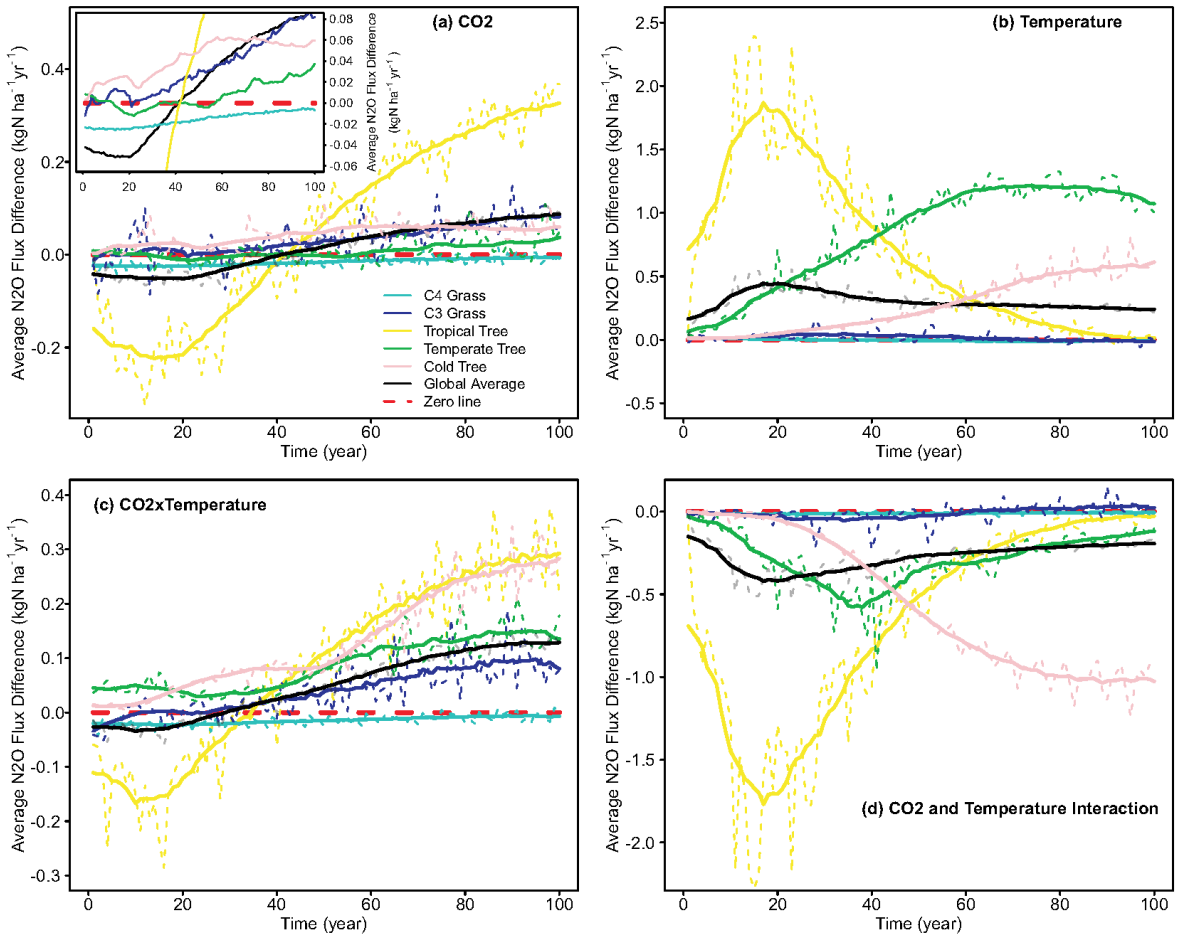


Figure 8. Soil N_2O emissions in response to step increases in atmospheric CO_2 and temperature. Panel (a) is the response to CO_2 fertilization alone, expressed as the difference between CO_2 increased run and the control run (CO2_FERT - CONTROL), the inset zooms into the y axis (flux difference) around zero; Panel (b) is the response to temperature increase alone (TEMP-CONTROL); Panel (c) is the combined response to both CO_2 enrichment and temperature rise (CO2_FERT×TEMP-CONTROL); and Panel (d) is the interactive effect of CO_2 and temperature responses, which is the difference between the combined (results from Panel (c)) and minus the individual responses (results from Panel (a) and (b)). Results are shown as annual values (thin dashed lines) and as running average with a moving window of 17 years (period of recycled climate forcing, thick solid lines) . The black lines represent the global average response. Coloured lines indicate responses for biome as represented by each plant functional type (PFT) considered in LM3V-N: C4 grass (cyan), C3 grass (blue), tropical forest (yellow), temperate deciduous forest (green) and cold evergreen forest (pink). Dashed red line represents the zero line.

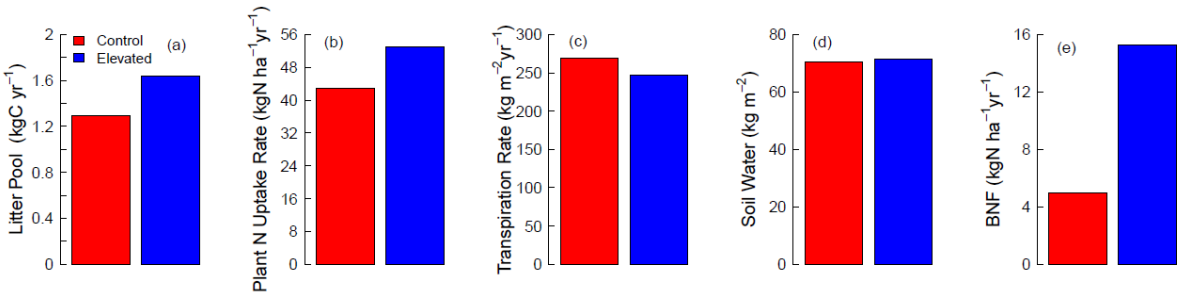
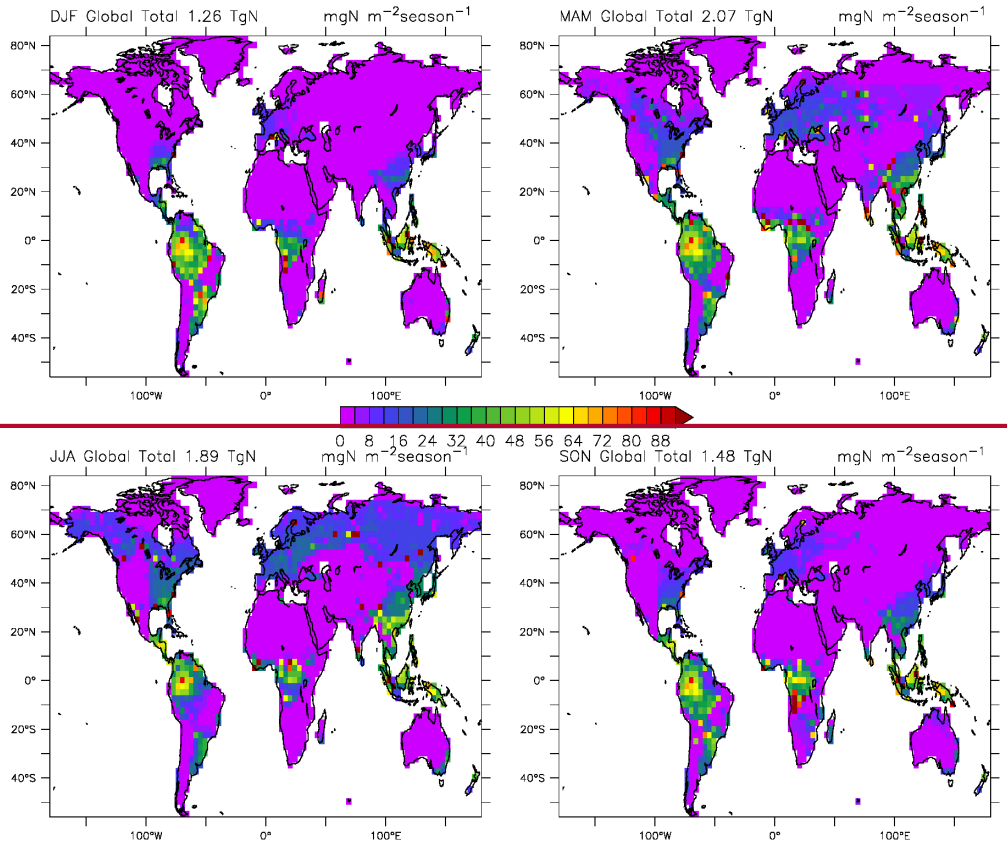


Figure 9. CO_2 fertilization effects (no temperature change) on litter pool size (Panel (a)), plant nitrogen uptake rate (Panel (b)), canopy transpiration rate (Panel (c)), soil water content in the root zone (Panel (d)) and biological nitrogen fixation (BNF) rate (Panel (e)). Shown are the 100-year average of global means (spatial) for control (284 ppm, red) and with elevated CO_2 (568 ppm, blue).

Table 1 Texture dependent parameter *k* estimated from Del Grosso et al. (2000)

Soil Texture	Coarse	Medium	Fine	Coarse medium	Coarse/ fine	Medium/ fine	Coarse/ medium/ fine	Organic
k	2	10	22	6	12	16	11	2



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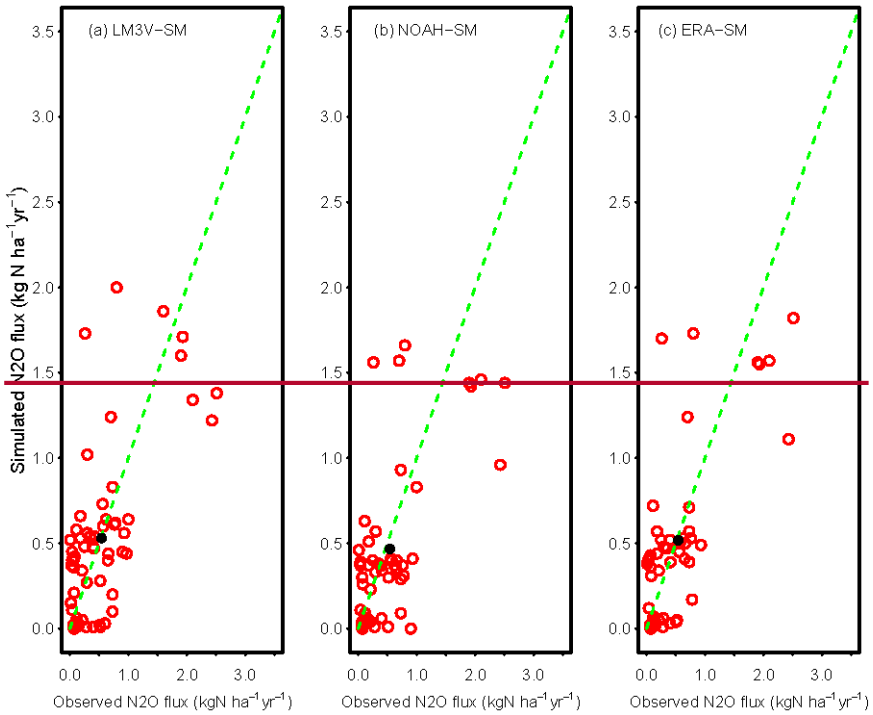
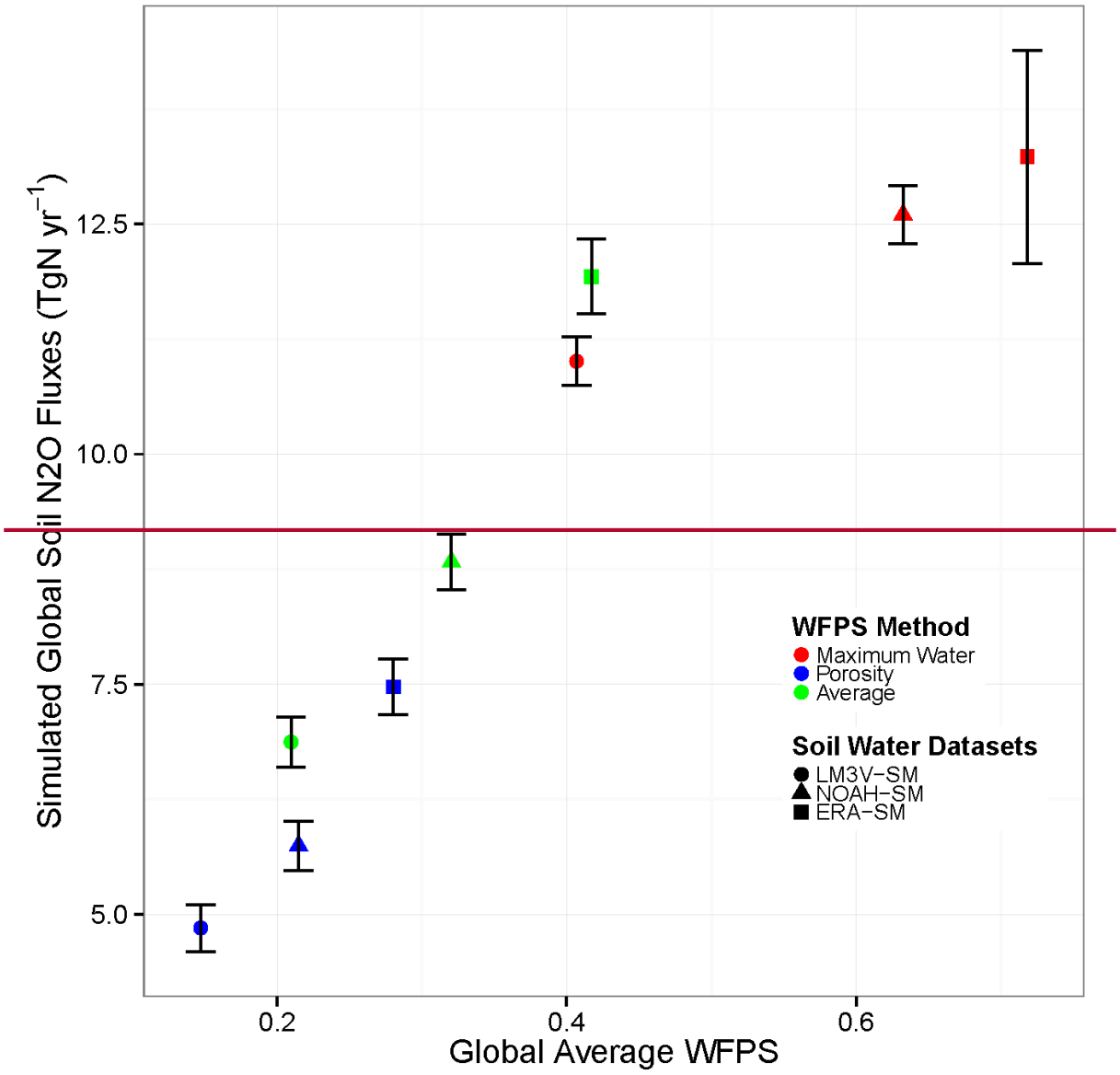


Figure 3. Observed vs. simulated annual N_2O emissions from natural soils. Dashed green lines are the 1:1 lines. The solid circles represent the overall means. Different panels represent simulations with different soil moisture data: (a) LM3V-SM (simulated by LM3V-N); (b) NOAH-SM (based on land surface model NOAH 3.3 in Global Land Data Assimilation System Version 2); and (c) ERA-SM (reanalysis data from ECMWF). Water filled pore space (WFPS) is calculated using the average of the one based on available water capacity and the one based on the total porosity (Method 3, see the main text for detailed description) for panel (a); and using the total porosity (Method 2) for panel (b) and (c).



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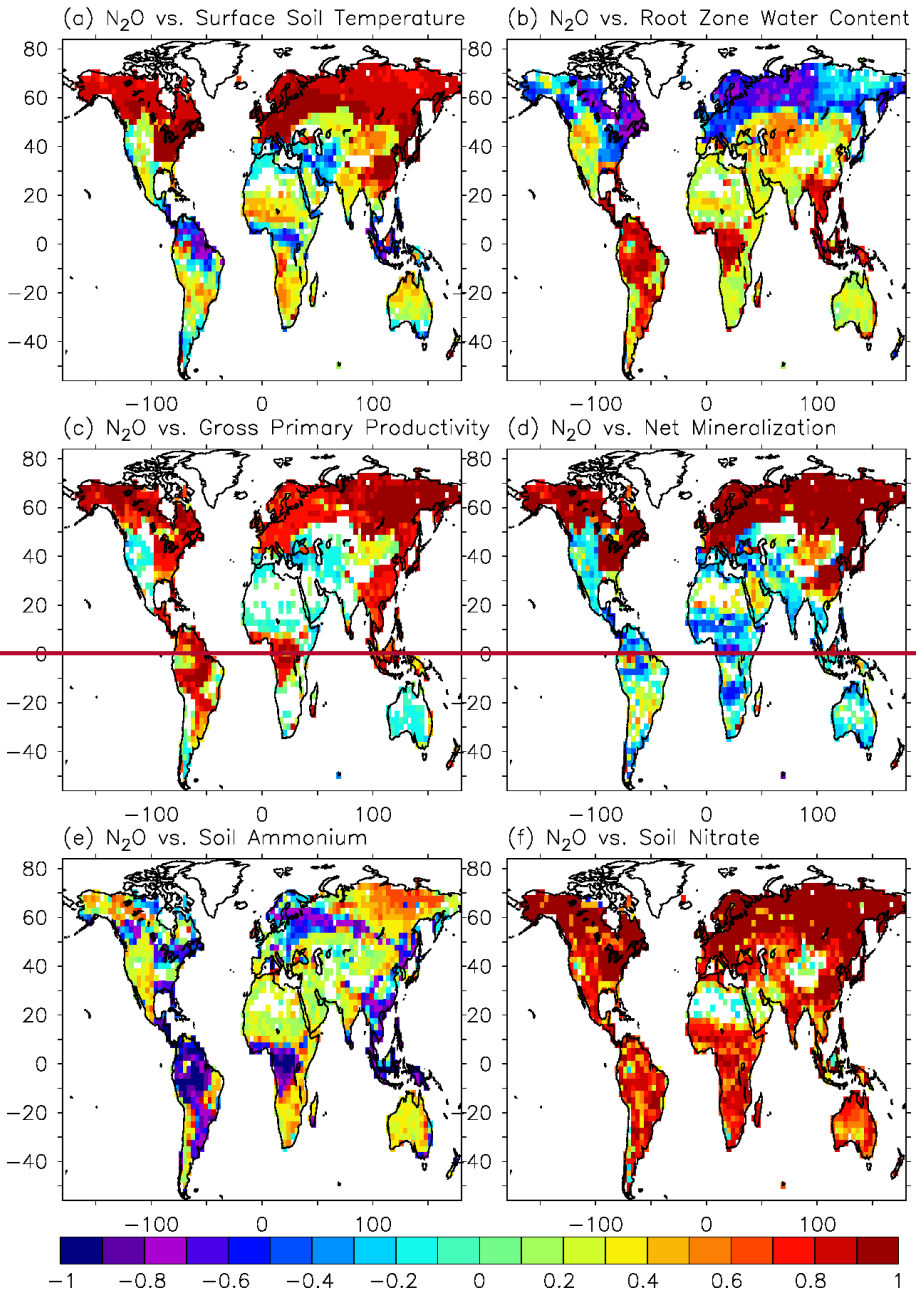
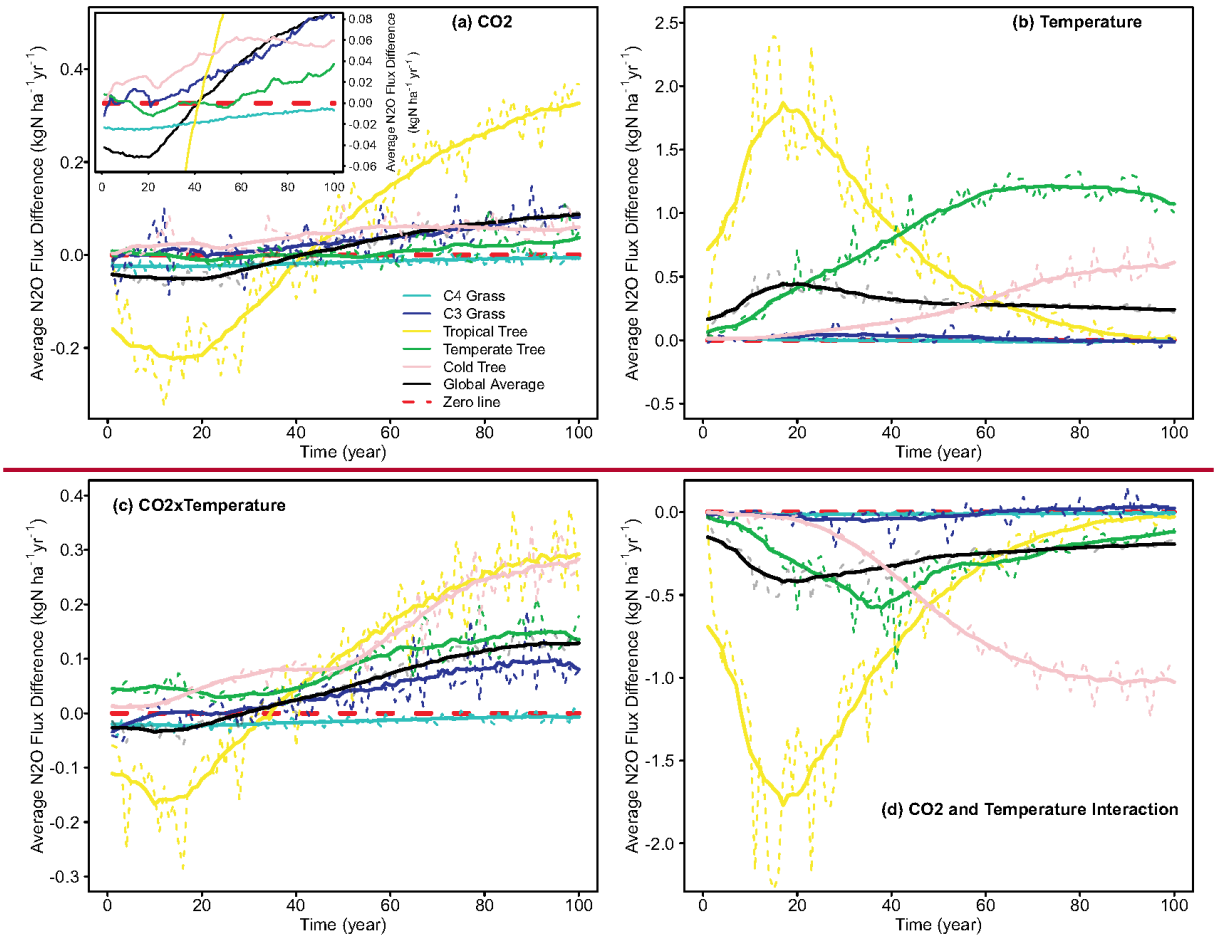


~~Figure 5. Temporal correlations between simulated monthly natural soil N_2O emissions and a) surface soil temperature, b) root zone water content, c) gross primary productivity, d) net mineralization, e) soil ammonium, and f) soil nitrate. White areas in panel a) to f) indicate locations either with no data or no significant ($\alpha >$) Pearson correlation coefficients.~~



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