Dear editor,

We received the comments from the executive editor and the two referees on our manuscript "Modeling the effects of litter stoichiometry and soil mineral N availability on soil organic matter formation" (gmd-2018-173). We are very grateful for their constructive comments and suggested amendments. We have carefully studied them, and revised our manuscript accordingly. As a consequence, our manuscript has been considerably improved.

The following part is our detailed responses to the comments from the executive editor and referees. Please note that the comments are in **bold** followed by our responses in regular text.

Sincerely,

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Response to the Executive editor of GMD

1. In my role as Executive editor of GMD, I would like to bring to your attention our Editorial version 1.1: http://www.geosci-model-dev.net/8/3487/2015/gmd-8-3487-2015.html. This highlights some requirements of papers published in GMD, which is also available on the GMD website in the 'Manuscript Types' section: http://www.geoscientific-model-development.net/submission/manuscript_types. html.

Thank you for this reminder. We have read the requirements of paper published in GMD carefully, and also adapted our manuscript accordingly to ensure it meets all the requirements of GMD. See below for details.

2. In particular, please note that for your paper, the following requirements have not been met in the Discussions paper: • "The main paper must give the model name and version number (or other unique identifier) in the title." • "If the model development relates to a single model then the model name and the version number must be included in the title of the paper. If the main intention of an article is to make a general (i.e. model independent) statement about the usefulness of a new development, but the usefulness is shown with the help of one specific model, the model name and version number must be stated in the title. The title could have a form such as, "Title outlining amazing generic advance: a case study with Model XXX (version Y)". In order to simplify reference to your developments, please add a model name (and/or its acronym) and a version number in the title of your article in your revised submission to GMD.

To fulfill these requirements, we have added the model name and version number in the title of our article. The original title has been changed from "Modeling the effects of litter stoichiometry and soil mineral N availability on soil organic matter formation"

to

"Modeling the effects of litter stoichiometry and soil mineral N availability on soil organic matter formation using CENTURY-CUE (v1.0)". (see lines 1-3)

Response to Referee #1

1.General Comments: Zhang and coauthors present a numerically tractable way to introduce variable carbon use efficiency (CUE) into a first-order litter decomposition model based on nitrogen availability. The paper is well written, with a very clean introduction that nicely summarizes relevant literature and concludes with a clear organization of the paper. Methods are adequately descriptive, results are clearly presented, and the discussion is on target (but see comment on N enrichment and litter decay below).

Thank you for your positive comments, and please see our responses to your concerns below.

2. Specific Comments: The approach outline here is nice, using short term experiments to calibrate the model and subsequently looking at the long-term dynamics. One concern, however, is that by using short term respiration rates from field and lab experiments to calibrate the variable CUE it is not clear if turnover coefficients that control litter mass loss are at all appropriate (more on this below).

Indeed, the litter turnover times have significant impacts on the fitted values of CUE. In our study, the turnover times for C pools are obtained from the ORCHIDEE-MICT that has good performances in reproducing observed organic carbon pools (v8.4.1, Guimberteau *et al*., 2018). However, we have calibrated the turnover times of the litter pools to the data of the incubation experiments. This calibration was necessary because the plant residues used in the incubation experiments of Recous *et al*. (1995) and Guenet *et al*. (2010) had been cut into fine fragments before being mixed with soil. It is known that the decomposability of litter is negatively correlated to its physical size (Tuomi *et al*., 2011). We further argue

that the mixing increases the accessibility of litter for microbes. Therefore, the turnover times of the incubated litter used in the experiments of Recous *et al.* (1995) and Guenet *et al.* (2010) should be shorter than the litter turnover times set in ORCHIDEE-MICT (24 days for metabolic litter and 89 days for structural litter), which are representative of to the turnover times of natural plant residues. In this study, we calibrated the turnover times of litter pools (metabolic and structural) based on the observed cumulative respired litter-C from all of the 14 incubation experiments using the M0 and M1 model (see Table A3 below).

We have added one paragraph to introduce the source of the SOC turnover times used in this study, and how we have calibrated the litter turnover times: "Note that the turnover times of SOM pools (active, slow and passive) used in this study are obtained from the ORCHIDEE-MICT (v8.4.1, Guimberteau *et al.*, 2018). The turnover times of litter pools (metabolic and structural), as well as the coefficient m_4 in Eq. (8) were optimized against the observed cumulative respired litter-C from all of the 14 incubation experiments using the M0 and M1 models (Table A3). A previous study has shown that litter decomposability is negatively correlated to its physical size (for example, Tuomi *et al.*, 2011). Therefore, the turnover times of the fine litter fragments used in the incubation experiments of Recous *et al.* (1995) and Guenet *et al.* (2010) are expected to be shorter than the values set in ORCHIDEE-MICT, which are representative of the turnover times of natural plant residues. In addition, the mixing of soil and litter particle in the incubation experiment likely enhances decomposition as spatial disconnection of decomposer and substrate, which can occur under natural soil conditions (Barnes *et al.*, 2012; Hewins *et al.*, 2013), is prevented. The calibrated turnover times of the metabolic and structural pools and the value of m_4 in Eq. (8) are 3.5 and 30 days and 0.5, respectively." (lines 465-483)

Table A3 List of parameters calibrated for two versions of the litter decomposition model (M0, M1): k_{litm} and k_{lits} are respectively the turnover rates of metabolic and structural litter pools, m_4 is the coefficient in Eq. (8), cue_{fit} is the optimized value of

CUE, m_1 and n_1 are the coefficients in Eq. (3), and m_2 is the coefficients in Eq. (5).

Version	CUE	$f(N_{min})$	Parameters
M0	Fixed	1	cue_{fit}, k_{litm}, k_{lits}, m_4
M1	Eqs.(2), (3)	Eq. (5)	m_1, n_1, m_2, k_{litm}, k_{lits}, m_4

(lines 1532-1536)

3. In section 2.5 it's a little unclear how the model and observations are disentangling background soil respiration from the litter respiration fluxes that are presumably being fit. Can this be clarified?

We have added some sentences to explain how the model and observations distinguish the litter- and SOC-derived CO_2.

For incubation experiments:

"To distinguish the litter- and SOC-derived CO_2 flux, Guenet *et al*. (2010) used straw from wheat grown under ^{13}C labeled CO_2 and they are therefore able to track the CO_2 coming from litter and the CO_2 coming from soil. In the experiments by Recous et al. (1995), litter-derived CO_2 flux is calculated as the difference in CO_2 flux between the incubation samples with both soil and litter, and the control samples without added litter." (lines 372-377)

For simulations:

"The observed cumulative respired litter-C (g C kg^{-1} soil) measured in the incubation experiments was used to calibrate the model parameter values. Moreover, to quantify the simulated CO_2 flux derived from the litter, we also performed a set of control simulations with only SOM (initial litter pools were set to 0 g kg^{-1} soil) using the four model versions. The simulated litter-derived CO_2 flux is calculated as the difference in CO_2 flux between the simulation with both litter and SOM inputs and the simulation with only SOM input." (lines 408-415)

4. I'm assuming there are no modifications to other CUE terms in CENTURY (between SOM pools), but this should be clarified.

We have added some sentences to clarify that only CUE for C transfers from litter pools to SOC pools were modified. Please see:

"Eqs. (2) and (3) were implemented in CENTURY to modify the originally fixed CUE_d (Fig. 1). With this change, the fractions of C from litter that remain in SOM are mediated by stoichiometric constraints and mineral N availability, at the expense of additional parameters to fit. The CUE_d for C transfers between SOC pools (active, slow and passive) are not modified." (lines 295-299)

5. Turnover times used in the model (e.g. tau_metabolic and tau_structural and well as the SOM turnover times listed in the github archive) are much larger than the litter turnover times used in CENTURY (Parton et al. 1988). This makes me wonder where the turnover parameterization here comes from? Addressing this concern is important since respiration rates are a product of turnover and CUE (given fixed initial pool sizes). Since the turnover times used here are much lower than in the CENTURY parameterization, the CUE will also have to be lower than if faster turnover times were used in the model. This is all fine, but should be made clearer in the text, which otherwise claims to be using the CENTURY approach.

The reviewer is correct; please see our response to Comment #2.

6. The maximum CUE allowed in the study seems quite high (0.8, Fig. 2). I'm assuming this assumption also causes the apparent high bias in CUE shown in Fig. 6? Is the model able to fit the data as well with a more reasonable upper limit for CUE (say 0.6), or is the high efficiency needed to capture results observed in the experiment?

We agree that CUE=0.8 is a relatively high value. While the CUEs of soil microbes are mostly concentrated between 0.4 and 0.6 (Manzoni et al., 2012), maximum values for reduced substrates are around 0.8 (Gommers et al., 1988), similar to maximum values also found in soils (Manzoni et al., 2012). Therefore, to allow the calibration procedure to cover a wide range of microbial CUEs, we set the maximum

CUE to 0.8. We have indicated the source reference of the maximum CUE in our manuscript. Please see:

"CUE_{max} (dimensionless) is the maximum CUE_d achieved when nutrients are not limiting, and it is set to 0.8 based on a synthesis of observed CUE of soil microbes (Manzoni *et al*., 2012)." (lines 264-266)

In addition, we also tested the performance of M1 model using a lower CUE_{max} of 0.6 as the referee suggests to be more reasonable. The result indicates that the optimized M1r is also able to capture the differences in respiration rates due to different C:N ratios of substrate and varying levels of mineral N availability across the 14 incubation experiments (Fig. R1b), although the RMSE (also AIC) of its estimation is slightly higher than that of M1 (Fig. R1c). The optimized function of *f(N_{min})* (Eq. 5) with a CUE_{max} of 0.6 is almost same to that with a CUE_{max} of 0.8 (Fig. R2b). But the optimized CUE_d function (Eq. 2) with a CUE_{max} of 0.6 is very different from that with a CUE_{max} of 0.8. When the CUE_{max} is set to 0.6, CUE_d increases very slowly with increasing soil mineral N concentration (Fig R2a), and shows almost no difference for litter with different qualities.
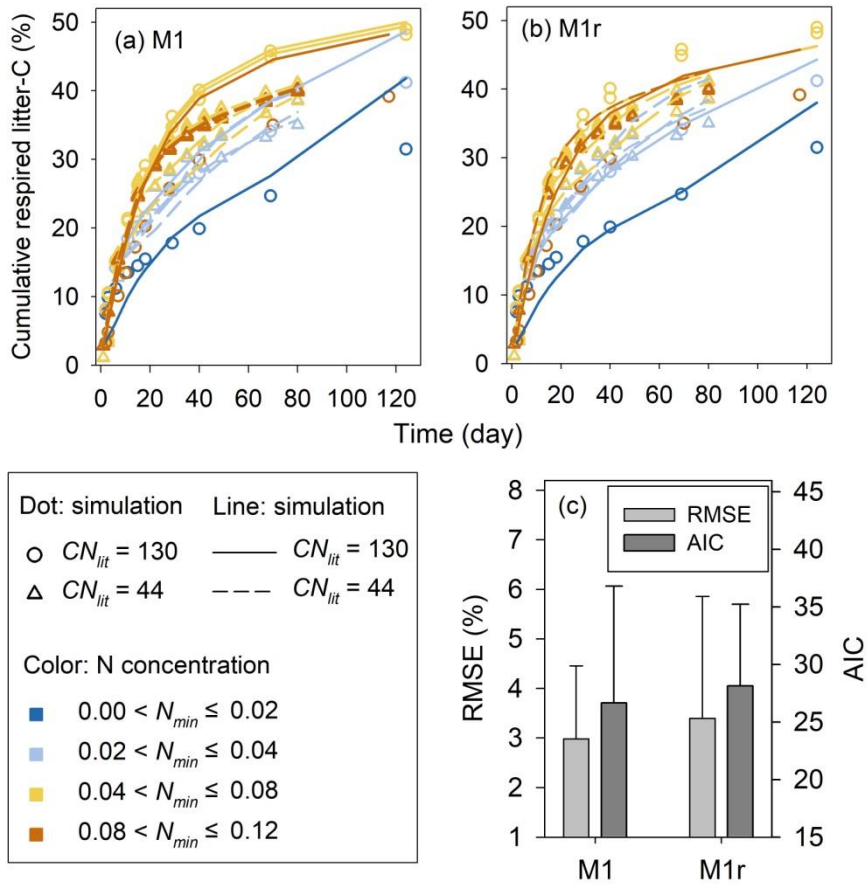
Figure R1 Comparison of simulated cumulative respired litter-C between models with CUE upper limit of 0.8 (M1) and 0.6 (M1r), respectively.
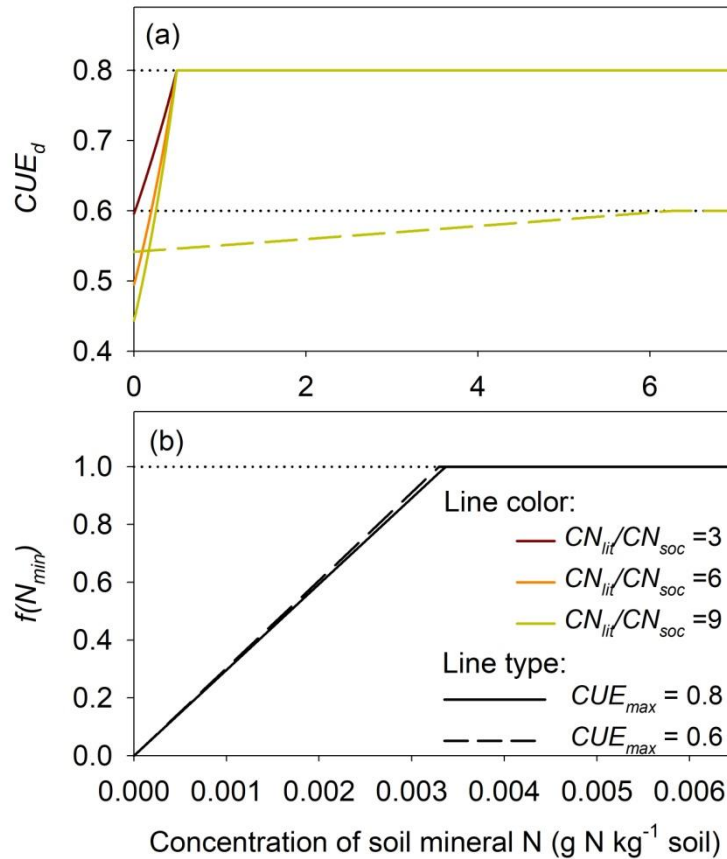
Figure R2. Change in the relations between carbon use efficiency (CUE_d, (a)) and N limitation factor ($f(N_{min})$, (b)), and mineral N concentration. Here the CUE_d and $f(N_{min})$ are calculated based on the optimized parameters when the upper limit of CUE is set to 0.8 (continuous line) and 0.6 (dashed line), respectively. CN_{lit} and CN_{SOM} are the C:N ratios of litter and SOM pools, respectively.

7. The main response of changes in CUE with litter quality seem to be opposite of what's expected. It seems like the authors expected to see a "decrease in CUEd with decreasing litter quality" (line 224), but instead report higher CUEd with the lower quality litter (line 363). Please explain how the parameterization let to this response and seems to contradict findings reported in Fig. 6.

There was a mistake in the text. We found an increase in CUE_d with declining litter quality. We revised the text:

"For very low quality litter with a C:N ratio of 130, the CUE_d in models M1 and M2 are <u>0.55 and 0.56</u>, respectively, which are <u>higher</u> than for better quality litter with C:N ratio of 44 (approximately <u>0.40 and 0.44</u> in M1 and M2, respectively)."

to

"For very low quality litter with a C:N ratio of 130, the CUE_d in models M1 and M2 are <u>0.40 and 0.44</u>, respectively, which are <u>lower</u> than for better quality litter with C:N ratio of 44 (approximately <u>0.55 and 0.56</u> in M1 and M2, respectively)." (lines 616-640)

8. Line 400. I agree, it's nice these parameters can be estimated, but the fit parameter values and their associated uncertainty are never communicated in the text. Can they be given in Table 1, or elsewhere in the manuscript? Similarly, does it make sense to include parameter values in Table A1?

We have added the parameter values and their associated uncertainties to the Table 1 in our manuscript. Please see:

"**Table 1** Optimized parameter values for the five versions of the litter decomposition model used in this study. cue_{fit} is the optimized value of CUE, m_1 and n_1 are the coefficients in Eq. (3), m_2 is the coefficient in Eq. (5), and m_3 is the coefficient in Eq. (6). Values in brackets following each parameter are the means ($\pm$ standard deviations) of the fitted parameter values based on "leave-one-out" cross-validation (see Section 2.5 for more details).

Version	CUE	$f(N_{min})$	Parameters
M0	Fixed	1	cue_{fit} (0.57±0.004)
M1	Eqs. (2), (3)	Eq. (5)	m_1 (0.61±0.34), n_1 (0.53±0.21), m_2 (297.4±38.0)
M2	Eqs. (2), (3)	1	m_1 (0.11±0.01), n_1 (1.96±0.13)
M3	Fixed	Eq. (5)	cue_{fit} (0.54±0.01), m_2 (396.9±23.6)
M4	Eqs.(2), (3)	Eq. (6)	m_1 (0.13±0.07), n_1 (1.91±0.37), m_3 (0.58±0.12)

" (lines 1409-1416)

9. The discussion is largely on target and I was very excited to see the authors try to take on results that generally show lower litter decomposition rates with N enrichment (e.g. Fog 1988, Knorr et al. 2005), line 415. What follows, however,

does not really conceptually address the apparent paradox of N additions, litter decay, and CUE. Instead the mathematical approach introduces new simulations and a new set of simulations (eq. 8, 9 & Fig. A5). Introducing new results like this in the discussion seems inappropriate for the journal. Instead it seems like these findings could be: (a) incorporated into the method and results; or (b) dropped from the manuscript. I would encourage the first option, but also ask the authors to more thoughtfully discuss how their results can inform larger questions about litter decay and N enrichment (Nave et al. 2009; Hobbie 2015; see also Wieder et al. 2015).

Thanks for your suggestion. We have moved the description of the alternative formulation for $f(N_{min})$ from the discussion section to the method section(see Section 2.4, lines: 326-340).

We added:

"The Model M4, which uses the alternative formulation for N constraints on litter decay (Eq. (6)), reproduces the different respiration rates of substrates with contrasting C:N ratios and at different levels of mineral N availability (Fig. A2), but with a slightly higher average RMSE of cumulative respired litter-C than model M1."in the Results section (lines 605-609)

We also added:

"In addition, the model M4, which is comparable to model M1 but uses an alternative formulation for N effects on the decomposition rate (Eq. (6)), performed slightly worse than model M1 (Fig. A2). Arguably, Eq. (6) represents the underlying mechanisms of N inhibition effects (Manzoni *et al*., 2009; Bonan *et al.*, 2013; Fujita *et al.*, 2014; Averill and Waring, 2018) better than Eq. (5) and due to the minor differences in RMSE and AIC (Figure A2b) between these formulations it can serve as an alternative to M1." in the Discussion section (lines 701-707)

Moreover, we have revised the discussions on the effects of N enrichment on litter respiration rate. The original sentences have been changed from "Moreover, describing N limitations on both the decomposition rate and flexible CUE$_d$ might allow our model to explain the observed diverse responses of litter respiration rate to

added mineral N in fertilization experiments (Hobbie and Vitousek, 2000; Guenet *et al.*, 2010; Janssens *et al.*, 2010). In these experiments, the net changes in respiration rate depend on the combined effects of added N on litter decay rate and CUE_d of the decayed litter (Fig. A4)."

to

"Our results indicate that the observed diversity of responses of litter respiration rate to mineral N additions (Hobbie and Vitousek, 2000; Guenet *et al.*, 2010; Janssens *et al.*, 2010) is likely due to the combined effects of changes in litter decay rate and CUE_d (Fig. A5). Thus, N addition effects can differ among fertilization experiments if litter quality and background N availability vary. In addition to altering litter decay rate and CUE_d, mineral N addition can induce abiotic formation of compounds that resist microbial attack, inhibit oxidative enzymes involved in lignin degradation, stimulate microbial biomass production early in decomposition, or lead to the accumulation of microbial residues that are resistant to decay (Fog, 1988; Hobbie, 2015). All these effects might decrease litter respiration rate by inhibiting the decomposition process, but have not been considered in our current model." (lines 708-728)

10. My final concern is somewhat subjective, but I argue that litter decomposition and SOM formation are not the same process. Throughout, however, the text (and especially the discussion) misleadingly conflates these two processes. While it's true that in first order models like CENTURY these processes are intimately linked, a growing body of literature highlights fundamental differences between processes controlling litter decay and SOM formation (see Lehmann and Kleber 2015, Sokol et al. 2018). Results shown in Fig. 7 are fine, but I would caution against linking these processes directly in the text.

This is also a good point. Indeed, litter decomposition and SOM formation are not the same processes, since SOM formation also involves stabilization processes. However, the first-order decomposition models like CENTURY have represented

these complicated processes in a very simple way, without explicit representation of the continuous transformation processes from decomposed litter to microbial productions and finally to stable SOM. According to your suggestion, we have revised our manuscript and deleted the sentences which might misleadingly conflate the litter decomposition and SOM formation processes. The major revision can be found from our response to your Comment #17 below. Please see lines 729-740 of the revised manuscript.

11. Technical corrections: Line 215, Don't 'microbes' include fungi and bacteria?

Line 215, Cleveland and Liptzin report microbial C:N = 8.6 (molar), so I'm assuming the 7.4 reported here on a mass basis, but this should be clarified in the text?

We have changed the original sentences from

"The C:N ratio of SOM (around 9:1 on a mass basis in CENTURY) is representative of the decomposer biomass, its value being between the C:N ratios of the two major group decomposers, soil microbes (7.4:1) (Cleveland and Liptzin, 2007)and soil fungi (13.4:1, Zhang *and* Elser, 2017)."

to

"The C:N ratio of SOM (around 9:1 on a mass basis in CENTURY) is representative of the decomposer biomass, its value being between the average C:N ratio of soil microbial communities including fungi and bacteria (7.4:1 in Cleveland and Liptzin, 2007) and the C:N ratio of soil fungi (13.4:1 in Zhang and Elser, 2017), which are probably largely responsible for fresh litter decomposition.". (lines 259-264)

12. Methods: It may be helpful to describe how the model handles partitioning of litter into metabolic and structural litter pools, and how the stoichiometry of these LIT pools changes with changes in litter quality (e.g., what are the donor pool C:N ratios if litterfall inputs have a C:N of 40 vs. 130)?

We have added a few sentences to introduce how the litter input is partitioned into

metabolic and structural pools, as well as how we set the C:N ratio of litter pools. Please see:

"Plant litter was firstly separated into metabolic and structural litter pools based on its lignin to C ratio (LC_{lit}, dimensionless). The fraction of metabolic litter-C (f_m, 0-1, dimensionless) is calculated by:

$$f_m = f_{max} - m_4 \times LC_{lit} \qquad (8)$$

where m_4 is a coefficient to be calibrated; f_{max}=0.85 is the maximum fraction of metabolic litter (i.e., the default value in CENTURY; Parton *et al*., 1988). The fraction of structural litter-C is thus 1- f_m. The C:N ratios of both metabolic and structural pools are assumed to be equal to the C:N ratio of litter input." (lines 398-406)

Note that, to avoid a double-consideration of the N content of litter input (that is to say the C:N ratio has been involved in the CUE formula), we just use the lignin content (Lignin:C) to calculate the fraction of metabolic litter. This is different from the algorithm used in the default CENTURY, which separates the litter inputs into metabolic and structural pools based on both lignin and N content.

13. Line 270 & 319, seems odd to cite a web site for a corporation selling composting material. A better choice may be Brovkin et al. 2012, who report litter quality estimates from the ART-DECO database, or work from the LIDET team (e.g. Harmon et al. 2009).

Thanks for your suggestion. We have changed the original sentence from "The C:N ratios of those corn residue and wheat straw span the range of litter C:N ratios among different ecosystems (Harmon *et al*., 2009; https://www.planetnatural.com/composting-101/making/c-n-ratio/)."

to

"The C:N ratios of those corn residue and wheat straw span the range of litter C:N ratios among different ecosystems (Harmon *et al*., 2009; Brovkin *et al*., 2012; Manzoni *et al*., 2010)." (lines 365-367)

and from "The assumed litter C:N ratios (CN_{lit}) of 10, 15, 30, 60, 120 and 200 span

the variation among most natural substrates and soil amendments from organic matter input in agriculture (Manzoni *et al.*, 2012; https://www.planetnatural.com/composting-101/making/c-n-ratio/).”

to

“The assumed litter C:N ratios (CN_{lit}) of 10, 15, 30, 60, 120 and 200 span the variation among most natural substrates and soil amendments from organic matter input in agriculture (Harmon *et al.*, 2009; Brovkin *et al.*, 2012; Manzoni *et al.*, 2010).” (lines 490-493)

14. Line 355, this statement isn't very obvious from Fig. 5b, in my estimation.

We have deleted the sentence “In addition, model M1 can also capture the temporal evolution of cumulative respired litter-C in different incubation experiments (Fig. 5b).”

15. Fig. 6. It's not really clear how the authors plot the C:N ratio of substrates : decomposers for a model that doesn't consider decomposers. I'm assuming this is the C:N ratio of donor (litter) / receiver (SOM pools; eq. 2)? Maybe this can be clarified in the figure caption? This is a fine assumption to make, although Cleveland and Liptzin (2007) found microbial C:N < soil C:N.

Sorry for the unclear explanation on the x-axis of Fig. 6. We have changed the original figure caption from “**Figure 6** Comparison of CUE_d (lines) predicted by Eq. (2) with parameter values (m2 = 0.54, n1 = 0.50) calibrated based on the incubation experiments (Table A2) of Recous *et al.* (1995) and Guenet *et al.* (2010) to observed CUE of terrestrial microorganisms along a gradient of CN_S/CN_D, where CN_D and CN_S are the C:N ratio of decomposers and their substrates, respectively. Gray dots are the estimated microbial CUE of litter decomposition in natural terrestrial ecosystems from Manzoni *et al.* (2017). Black squares are the microbial CUE measured via laboratory incubation experiments of Gilmour & Gilmour, (1985), Devêvre & Horwáth (2000) and Thiet *et al.* (2006). Error bars represent the standard deviations. N min (g N kg^{-1} soil) is the concentration of soil mineral N.”

to

"**Figure 6** Comparison of CUE_d (lines) predicted by Eq. (2) with parameter values ($m_2 = 0.54$, $n_1 = 0.50$) calibrated based on the incubation experiments (Table A2) of Recous et al. (1995) and Guenet et al. (2010)to observed CUE of terrestrial microorganisms along a gradient of CN_S/CN_D. For observed CUE (dots), CN_D and CN_S are the C:N ratio of decomposers and their substrates, respectively. For simulated CUE (lines), CN_S and CN_D correspond to the C:N ratio of donor (litter pool) and acceptor (the active SOM pool of the CENTURY), respectively. Gray dots are the estimated microbial CUE of litter decomposition in natural terrestrial ecosystems from Manzoni *et al.* (2017). Black squares are the microbial CUE measured via laboratory incubation experiments of Gilmour and Gilmour, (1985), Devêvre and Horwáth (2000) and Thiet *et al.*(2006). Error bars represent the standard deviations. N_{min} (g N kg^{-1} soil) is the concentration of soil mineral N." (lines 1494-1506)

16. How is Fig. A2 different from Fig. 5? Moreover, the caption in A2 doesn't seem to match the display item? (see also lines 374, 376).

The reviewer is correct: we have inserted a wrong figure as Fig. A2. Now we have corrected the error. Please see:
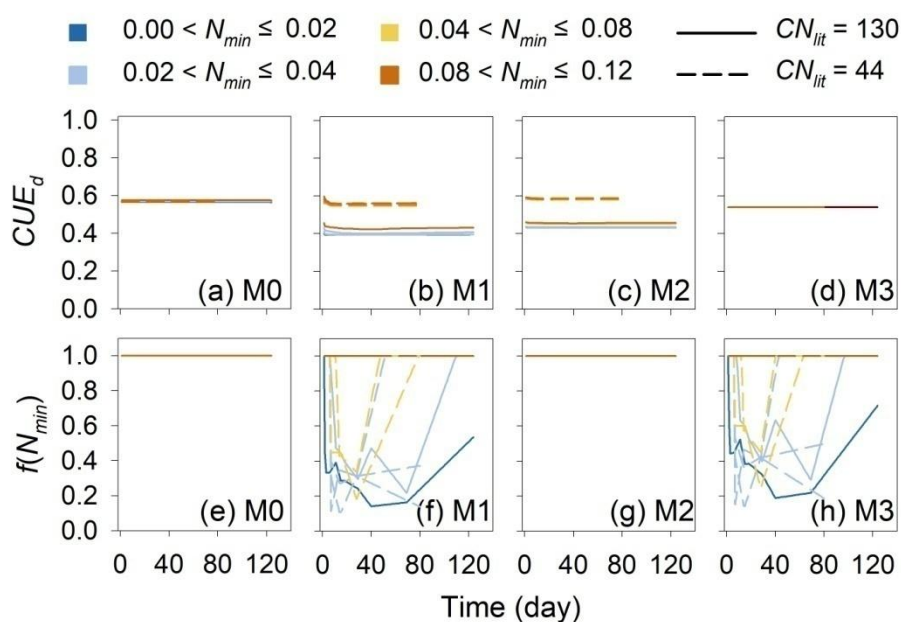
"

Figure A3 Dynamic of the simulated carbon use efficiency (*CUE*) and *f(N$_{min}$)* during the incubation experiments (Table A4). *CN$_{lit}$* is the C:N ratio of incubated litter, and *N$_{min}$* is the initial soil mineral N concentration (g N kg^{-1} soil). M0-M3 are the four models in Table 1. Here the simulation results of each model were calculated with parameters optimized based on all of the 14 samples of incubation experiments (Table A2)." (lines 1571-1577)

17. From line 480-506 on the discussion wanders well beyond the scope of results presented here. In particular, the emphasis on humic substances and litterfall driving SOM formation seems well out of line with contemporary thinking about factors controlling SOM stabilization (Lehmann and Kleber 2015). Moreover, the positive connection between CUE and steady-state SOM pools in first order models is well established (e.g. Frey et al. 2013). What's nice with the work presented here is the ability to link ideas about litter quality and SOM formation in ways that are consistent with theory about CUE and substrate quality (MEMs conceptual model, Cortufo et al. 2013) in a first order model. I'd encourage the authors more closely stick to interpreting the results presented with this work.

Thanks for your suggestion. We have revised the manuscript to make it more closely stick to interpreting the results presented with this work. The original sentences have been changed from "This study provides some insights on processes leading to increased SOM sequestration. Soil C sequestration plays a crucial role in food security and land CO_2 emission (Lal, 2004). The international initiative '4 per 1000' has been proposed to increase global SOM stock by 0.4% per year to compensate for anthropogenic CO_2 emissions (Baveye *et al.*, 2018). Transforming more plant litter into stable SOM (e.g. humic substances) has been suggested as an effective strategy to sequester more C in soil (Prescott, 2010). Our model results show a positive linear relationship between equilibrium SOC stock and CUE of decomposed litter (Fig. A4). This result can also be interpreted by calculating the analytical equilibrium SOC storage of a fully linear model including only one litter

pool and one SOC pool. In such a model, SOC receives C from the litter at a rate $CUE_d \times D$, where D is the litter decomposition rate, which equals to litterfall at steady state. SOC is lost via first order decay with a decay constant k. At steady state, input to and outputs from the SOC pool are equal and thus,

$$CUE_d \times D = k \times SOC \rightarrow SOC = CUE_d \frac{D}{k} \tag{11}$$

With a mean residence time of C in the SOC between 10 and 20 years and D approximated by litterfall (Table A4), SOC at equilibrium is predicted to scale linearly with CUE_d, with a slope approximately between 20 and 40, consistent with results in Fig. A4.

Therefore, litter quality needs to be controlled to maximize C sequestration in SOM pool (Eq. (2)). In line with previous studies (Prescott, 2010; Smith, 2016), our model predicts that adding N through fertilization and N-fixing plants will not only increase litter decay but also the fraction of litter-C being transformed into SOM and ultimately SOC stocks. However, application of mineral N fertilizer is associated with risk not considered here, like increasing land N_2O emission (Mosier and Kroeze, 2000; Kanter *et al*., 2016; Yi *et al*., 2017) and causing nitrate leaching which in turn can induce water pollution (Cao *et al*., 2006; Strokal *et al*., 2016). Due to the negative environmental impacts of mineral N addition, the use of N-rich litter substrates for increasing SOM is advised."

to

"This study provides insight on processes leading to increased SOM sequestration. Enhancing the efficiency at which plant residuals are transformed into stable SOM has been suggested as an effective strategy to sequester C in soil (Prescott, 2010; Cotrufo *et al*., 2013). Simulation results from our model suggest a positive linear relationship between equilibrium SOC stock and CUE of decomposed litter (Fig. A4), in line with the earlier findings with a similar model (for example Frey *et al*. 2013). In fact, with linear models such as CENTURY it can be shown that the steady state SOC scales linearly with CUE, different from nonlinear models predicting that higher CUE can trigger SOC loss (Allison *et al*., 2010). Our model goes beyond

earlier attempts (Bonan *et al.*, 2013; Fujita *et al.*, 2014; Averill and Waring, 2018) by adapting the optimal metabolic regulation hypothesis of Manzoni *et al.* (2017) to link CUE, litter quality and SOM formation in a process-oriented way." (lines 729-740)

18. Line 512, self-citations are nice, but it may also be worth referencing other modeling groups here?

We have changed the original sentence from "An increasing number of land surface models (e.g. ORCHIDEE-CNP, Goll *et al.*, 2017) have representations of the terrestrial N cycle."

to

"An increasing number of land surface models (Wang *et al.*, 2010; Zaehle *et al.*, 2014; Goll *et al.*, 2017) have representations of the terrestrial N cycle." (lines 1035-1036)

19. Line 516, didn't Bonan and others (2013, cited elsewhere in the text) already do this with CLM and CENTURY? Seems worth crediting work that's already been done along. these lines.

Although the constraint of soil mineral N availability on litter decomposition rate has been represented in some land surface and soil biogeochemical model (Bonan *et al.*, 2013; Fujita *et al.*, 2014; Averill and Waring, 2018), to our knowledge, none of these models have tested the links CUE to litter stoichiometry and soil nutrient availability. However, we acknowledge that other theoretical models have included this link (Schimel and Weintraub, 2003). The original sentence in our manuscript might have not given an accurate statement. We thus changed it from "By incorporating our litter decomposition formulation in these land surface models that simulate the dynamics of soil mineral N concentration, it will be possible to test and validate our developments with more extensive data from laboratory and field experiments."

to

"By incorporating our newly developed formulations of CUE_d and $f(N_{min})$ in these

land surface models that simulate the dynamics of soil mineral N concentration, it will be possible to test and validate our developments with more extensive data from laboratory and field experiments." (lines 1036-1040)

20. Line 538, the comparison with 'most large-scale litter decay models' was not made in this paper and I would remove this clause from the conclusion.

Thanks for your suggestion. We have removed this clause. The original sentence is changed from "Overall, the developed model captures the microbial mechanisms mediating litter stoichiometry and soil mineral N effects on litter decomposition and SOM formation <u>– representing an improvement over most existing large-scale litter decay models</u>."

to

"Overall, the developed model captures the microbial mechanisms mediating litter stoichiometry and soil mineral N effects on litter decomposition and SOM formation." (lines 1059-1066)

Response to Referee #2

1.This study adapted a conceptual formulation of CUEd based on assumption that litter decomposers optimally adjust their CUEd as a function of litter substrate C to nitrogen (N) stoichiometry. The new model algorithm was incorporated into CENTURY soil biogeochemical model and evaluated using data from laboratory litter incubation experiments. The results showed that new CUEd formulation with flexible CUE and effect of N availability to decay rate was able to reproduce differences in respiration rate of litter with contrasting C:N ratios and under different levels of mineral N availability. It is well-written, logically organized, and the figures and tables are appropriate.

Thanks for your positive comments.

2. Figure 1 seems too simple to include other major processes mentioned in the

method section. It should be considered to revise.

Thanks for your reminding. We have revised the Fig. 1 and checked the Method section to make sure that all important processes have been illustrated in this flowchart. Finally, we added the temperature (T (°C)) and soil moisture (SWC (%)) factors for scaling litter decay rate, as well as the inhibition effect of mineral N on litter decay rate ($f(N_{min})$). The original Fig. 1 has been changed from
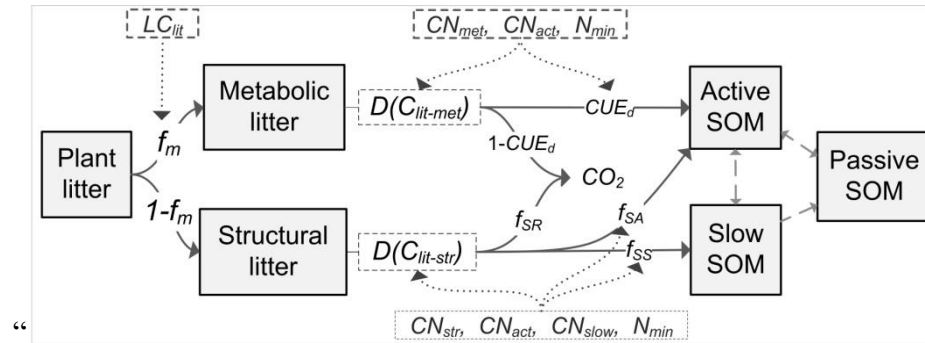
"

Figure 1 Schematic diagram of the C flows in the litter decay model used in this study. f_m is the fraction of metabolic compounds in plant litter. $D(C_{lit-met})$ and $D(C_{lit-str})$ are the decomposition rates (g C kg^{-1} day^{-1}) of metabolic or structural litter, respectively. LC_{lit} is the lignin:C ratio (on a mass basis) of plant litter; CN_{met}, CN_{str}, CN_{act}, and CN_{slow} are the C:N ratio of metabolic litter pool, structural litter pool, active SOM pool and slow SOM pool, respectively; N_{min} is the concentration of mineral N in solution (g N kg^{-1} soil); CUE_d is C use efficiency of the transformation from litter to soil organic matter (SOM); f_{SA}, f_{SS} and f_{SR} are the fractions of decomposed structural litter-C that is transferred to active SOM pool, slow SOM pool and released to atmosphere in forms of CO_2, respectively. As in the algorithms in CENTURY model (Parton *et al.*, 1988), here $f_{SA} = CUE_{d\_SA} \times (1-f_{lig})$, $f_{SS} = CUE_{d\_SS} \times f_{lig}$, $f_{SR} = 1-(f_{SA}+f_{SS})$, where f_{lig} is the lignin fraction (0–1, dimensionless) in the structural litter pool, and CUE_{d_SA} and CUE_{d_SS} are the CUE of C transformation from structural litter pool to active and slow SOM pool, respectively."

to

"

Figure 1 Schematic diagram of the C flows in the litter decay model used in this study. f_m is the fraction of metabolic compounds in plant litter. $D(C_{lit-met})$ and $D(C_{lit-str})$ are the decomposition rates (g C kg^{-1} day^{-1}) of metabolic or structural litter, respectively. LC_{lit} is the lignin:C ratio (on a mass basis) of plant litter; CN_{met}, CN_{str}, CN_{act}, and CN_{slow} are the C:N ratio of metabolic litter pool, structural litter pool, active SOM pool and slow SOM pool, respectively; N_{min} is the concentration of mineral N in solution (g N kg^{-1} soil); $\underline{f(N_{min})}$ is a factor reducing litter decay rate when soil mineral N availability is limiting; T (°C) and SWC (%) are temperature and soil water content, respectively; CUE_d is C use efficiency of the transformation from litter to soil organic matter (SOM); $\underline{CUE_{max}=0.8}$ is the maximum microbial CUE (dimensionless) when growth is limited by C from the organic substrate; f_{SA}, f_{SS} and f_{SR} are the fractions of decomposed structural litter-C that is transferred to active SOM pool, slow SOM pool and released to atmosphere in forms of CO_2, respectively. As in the algorithms in CENTURY model (Parton et al., 1988), here $f_{SA}=CUE_{d\_SA}\times(1-f_{lig})$, $f_{SS}=CUE_{d\_SS}\times f_{lig}$, $f_{SR}=1-(f_{SA}+f_{SS})$, where f_{lig} is the lignin fraction (0–1, dimensionless) in the structural litter pool, and CUE_{d_SA} and CUE_{d_SS} are the CUE of C transformation from structural litter pool to active and slow SOM pool, respectively." (lines 1428-1445)

3. As the CUEd was defined as a fraction of it is respired to the atmosphere and the remaining fraction (Line 159-160), it is not correct to use 1-CUEd to simulate CO2 emission in Fig. 1.

Microbial carbon use efficiency (CUE),defined as the ratio of microbial biomass production to material uptake from substrates (lines 68-69). In our study, the CUE of

decayed litter-C ($D_{(C\text{-}lit)}$) is defined as the ratio of C that is transferred into SOC pool (CUE×$D_{(C\text{-}lit)}$)to the total decayed litter-C. Therefore, the remaining fraction ((1-CUE)×$D_{(C\text{-}lit)}$) is respired to the atmosphere as $CO2$. To explain the definition of CUE more explicitly, we have changed the original sentence from "When C is being transferred between pools, a fraction of it is respired to the atmosphere and the remaining fraction (CUE_d conceptually equal to microbial CUE) enters the acceptor pool."

to

"When litter is being decomposed, a fraction of the decomposed C is respired to the atmosphere and the remaining fraction (CUE_d conceptually equal to microbial CUE) enters the acceptor SOM pool." (lines 196-199)

4. Equ (4) is important for this study, which has been used to develop one of model simulations (i.e. M1). However what is the fundamental assumption for adding N effects in the Equ (4)? N mineralization is accompanied with carbon decomposition. So, why use N availability to limit litter decay?

Biomass of microbes is stoichiometrically constrained. When the supply of N from substrates is lower than the demand of microbes to fulfill their specific stoichiometric C:N ratio, microbes will utilize the mineral N (immobilization). Thus low availability of mineral N can limit microbial activity, and in turn litter decay rate. There is no explicit representation of microbial growth in CENTURY model. But the C:N ratio of SOM pool is assumed to be same to that of the microbial biomass. The mineralized N accompanying with litter decomposition will preferentially enter SOM pool to fulfill the SOM C:N ratio. When the N supply from decomposed litter is lower than the demand of newly formed SOM, soil mineral N will be immobilized. Therefore when soil mineral N concentration is very low and the immobilized N cannot meet the N demand of SOM, then the mineral N becomes a constraint factor of litter decomposition rate.

We have provided a brief introduction on the fundamental assumption for adding the mineral N factor in Eq. 4, and it can be find from:"Microbial biomass is nearly

homeostatic (Cleveland and Liptzin, 2007; Franklin *et al.*, 2011; Allison, 2012).

When the supply of N from substrates is lower than the demand of microbes to fulfill

their specific stoichiometric C:N ratio, microbes will utilize mineral N

(immobilization) (Manzoni *et al.*, 2012). Thus, low availability of mineral N can

limit microbial activity, and in turn litter decay rate (Manzoni and Porporato 2009;

Fujita *et al.*, 2014)." (lines 130-135)

and

"The C:N ratio of SOM (around 9:1 on a mass basis in CENTURY) is representative

of the decomposer biomass, its value being between the average C:N ratio of soil

microbial communities including fungi and bacteria (7.4:1 in Cleveland and Liptzin,

2007) and the C:N ratio of soil fungi (13.4:1 in Zhang and Elser, 2017), which are

probably largely responsible for fresh litter decomposition." (lines 259-264)

5. Line 71: need reference here.

We have added references for our statement: "During litter decomposition, only a

part of the decomposed litter-C is being transferred into SOM, while the remaining C

is being released as CO_2 to the atmosphere by microbial respiration (Campbell and

Paustian, 2015; Cotrufo *et al*., 2015)." (lines 73-76)

6. Line 212: typo "The The C:N ratio"

Sorry for the mistake. We have changed the original sentence from "The The C:N

ratio of SOM (around 9:1 on a mass basis in CENTURY) is representative of the

decomposer biomass, its value being close to the observed average C:N ratio of soil

microbes (7.4:1 in Cleveland and Liptzin, 2007 and 13.4:1for soil fungi in Zhang *and*

Elser, 2017)."

to

"The C:N ratio of SOM (around 9:1 on a mass basis in CENTURY) is representative

of the decomposer biomass, its value being between the average C:N ratio of soil

microbial communities including fungi and bacteria (7.4:1 in Cleveland and Liptzin,

2007) and the C:N ratio of soil fungi (13.4:1 in Zhang and Elser, 2017), which are

probably largely responsible for fresh litter decomposition." (lines 259-264)

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Modeling the effects of litter stoichiometry and soil mineral N availability on soil organic matter formation using CENTURY-CUE (v1.0)

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Type: primary research

Abstract

Microbial decomposition of plant litter is a crucial process for the land carbon (C) cycle, as it directly controls the partitioning of litter-C between CO_2 released to the atmosphere versus the formation of new soil organic matter (SOM). Land surface models used to study the C cycle rarely considered flexibility in the decomposer C use efficiency (CUE_d) defined by the fraction of decomposed litter-C that is retained as SOM (as opposed to be respired). In this study, we adapted a conceptual formulation of CUE_d based on assumption that litter decomposers optimally adjust their CUE_d as a function of litter substrate C to nitrogen (N) stoichiometry to maximize their growth rates. This formulation was incorporated into the widely used CENTURY soil biogeochemical model and evaluated based on data from laboratory litter incubation experiments. Results indicated that the CENTURY model with new CUE_d formulation was able to reproduce differences in respiration rate of litter with contrasting C:N ratios and under different levels of mineral N availability, whereas the default model with fixed CUE_d could not. Using the model with flexible CUE_d, we also illustrated that litter quality affected the long-term SOM formation. Litter with a small C:N ratio tended to form a larger SOM pool than litter with larger C:N ratios, as it could be more efficiently incorporated into SOM by microorganisms. This study provided a simple but effective formulation to quantify the effect of varying litter quality (N content) on SOM formation across temporal scales. Optimality theory appears to be suitable to predict complex processes of litter decomposition into soil C, and to quantify how plant residues and manure can be harnessed to improve soil C sequestration for climate mitigation.

Keywords: microbial carbon use efficiency, litter decomposition, litter stoichiometry, soil organic matter, litter decay model, nitrogen

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1 Introduction

Plant litter decomposition plays a key role in global carbon (C) cycle, thus needs to be well represented in land surface models. The decomposition and transformation processes of plant litter control the formation of soil organic matter (SOM) (Prescott, 2010; Schmidt *et al.*, 2011; Walela *et al.*, 2014; Cotrufo *et al.*, 2015) and associate immobilization and mineralization of essential plant nutrients (Moorhead and Sinsabaugh, 2006; Parton *et al.*, 2007; Manzoni *et al.*, 2008; Manzoni and Porporato, 2009). Hence, a reliable litter decay model is necessary for estimating soil C balance and turnover of ecosystem C (Allison, 2012; Bonan *et al.*, 2013; Wieder *et al.*, 2013; Campbell and Paustian, 2015). In particular, a realistic representation of litter decomposition in land surface models is helpful to decrease the uncertainties in predicted effects of climate change and anthropogenic management on ecosystems (Gholz *et al.*, 2000; Campbell and Paustian, 2015; Luo *et al.*, 2016). As litter decomposition is a very complex process determined by climate (e.g. temperature and moisture), litter quality (e.g. nitrogen (N) concentration), soil nutrients and the physiological characteristics of microorganisms (Lekkerkerk *et al.*, 1990; Prescott, 2010; Manzoni *et al.*, 2012; Frey *et al.*, 2013; Sinsabaugh *et al.*, 2013; García-Palacios *et al.*, 2016), there remain large uncertainties in existing litter decay models (Zhang *et al.*, 2008; Bonan *et al.*, 2013; Campbell and Paustian, 2015). Many litter decay models, especially those incorporated in global land surface models, have ignored stoichiometric constraints to microbial processes (Bonan *et al.*, 2013; Cotrufo *et al.*, 2013; Wieder *et al.*, 2013; Wieder *et al.*, 2014).

Microbial carbon use efficiency (CUE), defined as the ratio of microbial biomass production to material uptake from substrates (Lekkerkerk *et al.*, 1990; Manzoni *et al.*, 2012), is an important parameter constraining litter decay, but it has rarely been represented as a flexible quantity in land surface models. During litter decomposition, only a part of the decomposed litter-C is being transferred into SOM, while the remaining C is being released as CO_2 to the atmosphere by microbial respiration (Campbell and Paustian, 2015; Cotrufo *et al.*, 2015). While CUE is a

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84 physiological property of each decomposer community, it also determines the

85 ecosystem-level efficiency at which litter C is transferred into SOM a step further from

86 simple microbial incorporation. We denote this efficiency as carbon use efficiency of

87 litter decomposition (CUE_d). With higher CUE_d, more plant-produced litter is

88 transformed biologically into SOM, and soil C storage can reach higher values (Six *et*

89 *al.*, 2006; Sinsabaugh *et al.*, 2013). In most existing soil biogeochemical models,

90 CUE_d is assumed to be same as microbial CUE and considered as a fixed parameter.

91 The Verberne model (Verberne *et al.*, 1990) assumes for instance $CUE_d \approx 0.25$. In the

92 Yasso model (Liski *et al.*, 2005), the CUE_d is set to 0.2. The CENTURY model sets

93 the CUE_d for decomposition of surface and belowground metabolic litter to 0.55 and

94 0.45, respectively (Parton *et al.*, 1988). In Daisy (Hansen *et al.*, 1991), NCSOIL

95 (Molina *et al.*, 1983) and ICBM (Kätterer and Andrén, 2001), $CUE_d = 0.6$ for the labile

96 litter pools and takes a lower value for recalcitrant substrates. Only a few models

97 account for variable CUE, letting it vary in response to substrate stoichiometry

98 (Schimel and Weintraub, 2003) or temperature (Allison *et al.*, 2010).

99 The increasing evidence for a variable microbial CUE leads to a conceptual

100 CUE model which can explain trends in CUE of microorganisms along stoichiometric

101 gradients (Manzoni *et al.*, 2017). The values of CUE_d used in existing litter decay

102 models are mostly derived from CUE obtained in laboratory studies on microbial

103 physiology or limited observations in certain ecosystems, thus show large variations

104 without a dynamic link to environmental conditions (Parton *et al.*, 1988; Verberne *et*

105 *al.*, 1990; Hansen *et al.*, 1991; Liski *et al.*, 2005; Manzoni *et al.*, 2012). Recent studies

106 (Manzoni *et al.*, 2008, 2012) suggested that the microbial CUE in terrestrial

107 ecosystems ranges from less than 0.1 for wood decomposers to about 0.5 for

108 decomposition of N-rich and high-quality litter. To explain those differences, Manzoni

109 *et al.* (2017) proposed a conceptual model of microbial CUE based on the assumption

110 that decomposers adapt their metabolism (and hence CUE) to maximize their growth

111 rate. This model based on optimality theory links CUE to substrate and decomposers

112 stoichiometry, where the optimal CUE decreases with increasing substrate

113 C-to-nutrient ratio, and increases with soil nutrient availability. The predictions of this

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124 theoretical model have been verified by empirical evidence from CUE estimates for

125 different microorganisms in both aquatic and terrestrial ecosystems (Manzoni *et al.*,

126 2017).

127 Besides variable CUE_d, many previous studies have also indicated the

128 necessity for litter decomposition models to consider soil mineral N availability as a

129 driver of litter decomposition rates, in particular under low N availability (Wieder *et*

130 *al.*, 2015; Luo *et al.*, 2016; Averill and Waring, 2018). Microbial biomass nearly

131 homeostatic (Cleveland and Liptzin, 2007; Franklin *et al.*, 2011; Allison, 2012). When

132 the supply of N from substrates is lower than the demand of microbes to fulfill their

133 specific stoichiometric C:N ratio, microbes will utilize mineral N (immobilization)

134 (Manzoni *et al.*, 2012).Thus, low availability of mineral N can limit microbial activity,

135 and in turn litter decay rate (Manzoni and Porporato 2009; Fujita *et al.*, 2014).

136 Although there are fertilization experiments reporting insignificant or even negative

137 impacts of added N on litter decay rate (Fog, 1988; Hobbie and Vitousek, 2000; Finn

138 *et al.*, 2015), many incubation experiments showed a significant decrease of litter

139 decomposition rate with declining mineral N availability (Recous *et al.*, 1995; Hobbie

140 and Vitousek, 2000; Guenet *et al.*, 2010). Moreover, recent modeling studies have

141 indicated that including the limiting effect of low mineral N on decomposition

142 improved predictions of C and N fluxes (Bonan *et al.*, 2013; Fujita *et al.*, 2014).

143 Therefore, soil mineral N can alter litter C flux by affecting both the litter decay rate

144 and the partition of decayed litter-C (via flexible CUE_d).

145 Some detailed microbial decomposition models actually have included

146 variable microbial CUE and the limitation of low mineral N availability on litter

147 decay rate (Ingwersen *et al.*, 2008; Pagel *et al.*, 2013; Campbell *et al.*, 2016; Huang *et*

148 *al.*, 2018); however, the parameterization and evaluation of these models pose

149 significant challenges due to their complexity and limited verification data (Wieder *et*

150 *al.*, 2014; Campbell and Paustian, 2015). There is still scope for implementing the

151 effects of litter stoichiometry and soil mineral N availability on litter decomposition in

152 litter decay models with more generalizable structure. In particular, it is important to

153 test the role of these effects in models that have already been incorporated into land

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surface model for long-term and large-scale applications (e.g. CENTURY, Parton *et al.*, 1988). In this study, we incorporated flexible CUE_d based on substrate C:N ratios and mineral N limitations into a soil biogeochemical model based on the CENTURY equations to simulate the decomposition and transfer processes of litter-C. The study was organized as follows. First, the new model was calibrated and tested against data from laboratory litter incubation experiments for its ability to capture the effect of variable litter quality and soil mineral N on litter respiration rates (short-term simulations). Second, the model parameterized assuming flexible CUE_d and mineral N limitations was used to explore the consequences of such stoichiometric constraints on the production of soil organic carbon (SOC) (long-term simulations). With these two modeling analyses, we aimed at linking stoichiometric constraints acting on short-term (months to years) decomposition dynamics to their consequences on SOC accumulation occurring at decadal to centennial time scales.

2 Materials and methods

2.1 The CENTURY decomposition model

The basis of the litter decay model used in this study is the CENTURY model (Fig. 1), a first-order decay model that describes decomposition as a function of substrate availability and quality, clay content, soil moisture and soil temperature (Parton *et al.*, 1988). Most land surface models (e.g. Kucharik *et al.*, 2000; Sitch *et al.*, 2003; Krinner *et al.*, 2005) adopted a similar structure to simulate the litter and soil biogeochemical processes. Dead organic matter in CENTURY is separated into structural and metabolic litter and three SOM pools (active, slow, passive) with different turnover times. There is no explicit representation of microbial biomass in CENTURY, instead the biomass of microbes is assumed to be in equilibrium with labile SOM and thus implicitly included in the active SOM pool. When litter is being decomposed, a fraction of the decomposed C is respired to the atmosphere and the remaining fraction (CUE_d conceptually equal to microbial CUE) enters the acceptor SOM pool. Three of such fractions are defined to characterize the transfer of C from

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litter to SOM: CUE_{ma} for transfer of the metabolic litter to the active SOM pool, and

CUE_{sa} and CUE_{ss} for transfer of structural litter to active and slow SOM pools,

respectively (Fig. 1). These fractions are set to be time invariant in the original version

of CENTURY, so that a fixed fraction of decomposed C is retained in the acceptor

pool regardless of environmental conditions and changes in the quality of the donor

pool. The N flows in CENTURY follow the C flows and are equal to the product of C

flow by the N:C ratio of the acceptor SOM pool. N mineralization is defined as the

difference between N obtained from the donor pools and N stoichiometric demand of

the acceptor pool (Parton *et al*., 1988; Metherell *et al*., 1993). In this way, net N

mineralization occurs when the donor pool has low C:N ratio, but N is immobilized

(taken up by microbes) when the donor pool has high C:N ratio.

2.2 Optimal CUE

To quantify how microbial CUE varies along gradients of nutrient

availability, it can be hypothesized that microorganisms maximize their growth rate,

and hence their ecological competitiveness, by adapting resource (C and nutrients) use

efficiencies. This follows the growth maximization hypothesis (Mooshammer *et al*.,

2014; Manzoni *et al*., 2017). Based on this hypothesis, Manzoni *et al*. (2017)

formulated a theoretical model expressing microbial CUE as a function of the

stoichiometric difference between decomposers and their substrate. The CUE for

which growth rate is maximized is the optimal CUE (CUE_{opt}) given by:

$$CUE_{opt} = CUE_{max} \times \min\left[1, \frac{CN_D}{CUE_{max}} \times \left(\frac{1}{CN_S} + \frac{I_N}{U_0}\right)\right] \tag{1}$$

where CUE_{max} is the maximum microbial CUE (dimensionless) when growth is

limited by C from the organic substrate. CN_D and CN_S are the C:N ratio (in mass,

dimensionless) of decomposer and their substrate, respectively. Although Manzoni *et

al*. (2017) indicated that mineral phosphorus (P) could also affect optimal CUE we

only considered N as a limiting nutrient. I_N (g N kg^{-1} soil) is the maximum rate at

which mineral N can be taken up by microbes, and U_0 (g C kg^{-1} soil) is the C-limited

uptake rate (corresponding to the decomposition rate at optimal mineral N

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concentration). When litter C:N is low or soil mineral N is in excess, the second term

in the minimum function (Eq.(1)) is higher than one, and $CUE_{opt} = CUE_{max}$ (C limited

conditions, as in nutrient-rich litter). In contrast, when mineral N is scarce, CUE_{opt}

decreases with increasing substrate C:N ratio (N limited conditions, N-poor litter).

Lack of N in the organic substrates can be compensated by mineral N being

immobilized by microorganisms from the soil solution. Immobilization meets the

nutrient demands as long as it is lower than the maximum supply rate I_N, at which

point microbial CUE starts being down regulated. Thus, for any given C:N ratio in the

substrate, CUE_{opt} increases with inorganic N concentration in the soil solution until

CUE_{max} is reached. It should also be noted that Eq. (1) is interpreted at the microbial

community scale, not for individual organisms.

2.3 Adaption of the optimal CUE model in the CENTURY model

      CUE of decomposition (CUE$_d$) is also assumed to be equivalent to microbial

CUE in this study. Then we followed the theory from Manzoni *et al.* (2017) (Eq. (1))

to parameterize CUE$_d$ during litter decomposition into CENTURY (Fig.1). Due to the

implicit representation of microbial growth in CENTURY, we replaced the original

optimality CUE model (Eq. (1)) by a simpler equationthat involves the C:N ratios of

the donor and acceptor pools, rather than microbial C:N ratios:

$$CUE_{opt} = CUE_{max} \times \min\left[1, \left(\frac{CN_{lit}}{CN_{SOM}}\right)^a\right] \tag{2}$$

where CN_{lit} and CN_{SOM} are the C:N ratio (dimensionless) of litter (metabolic or

structural) and SOM pools (active, slow or passive), respectively. The C:N ratio of

SOM (around 9:1 on a mass basis in CENTURY) is representative of the decomposer

biomass, its value being between the average C:N ratio of soil microbial communities

including fungi and bacteria (7.4:1 in Cleveland and Liptzin, 2007) and the C:N ratio

of soil fungi (13.4:1 in Zhang and Elser, 2017), which are probably largely

responsible for fresh litter decomposition. CUE_{max} (dimensionless) is the maximum

CUE$_d$ achieved when nutrients are not limiting, and it is set to 0.8 based on a

synthesis of observed CUE of soil microbes (Manzoni *et al.*, 2012). The exponent a (g

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N kg^{-1} soil) captures the effect of mineral N uptake by microbes on CUE$_d$. Because CUE$_d$ is expected to increase with mineral N availability (Eq. (1)), a is assumed to be a linear function of the mineral N concentration (N_{min}, g N kg^{-1} soil):

$$a = m_1 \times (N_{min} - n_1) \tag{3}$$

where m_1 (kg g^{-1} N) and n_1 (g N kg^{-1} soil) are two coefficients that need to be calibrated. Eqs. (2) and (3) modulate the decrease in CUE$_d$ with decreasing litter quality when mineral N availability changes– the exponent a increases with increasing mineral N availability, causing an increase in CUE$_d$ at any given litter C:N ratio. Hence, increasing a mimics an increase in I_N in Eq. (1). Fig. 2a illustrates how CUE$_d$ from Eq. (2) varies as a function of mineral N concentration, for different values of litter C:N.

Eqs. (2) and (3) were implemented in CENTURY to modify the originally fixed CUE$_d$ (Fig. 1). With this change, the fractions of C from litter that remain in SOM are mediated by stoichiometric constraints and mineral N availability, at the expense of additional parameters to fit. The CUE$_d$ for C transfers between SOC pools (active, slow and passive) are not modified.

2.4 Constraint of soil nutrient availability on litter decomposition rate

CENTURY is a first-order decay model in which decomposition rates of metabolic and structural litter are modulated by scaling factors of soil temperature ($f(tem)$) and moisture ($f(water)$) (Parton $et\ al.$, 1988). Here, we introduced an additional mineral N scaling factor ($f(N_{min})$, 0–1, dimensionless) to account for the limitation imposed by low mineral N availability on litter decay rate ($D(C_{lit})$).

$$D(C_{lit}) = C_{lit} \times k \times f(tem) \times f(water) \times f(N_{min}) \tag{4}$$

where C_{lit} is the C (g C kg^{-1} soil) in litter pool (metabolic or structural) and k is the potential maximum turnover rate (day^{-1}) at optimal soil temperature, moisture and nutrient conditions.

In this study, we assumed that the scaling factor of mineral N increases linearly with increasing soil mineral N concentration (N_{min}, Eq. (5)) below a threshold value of $1/m_2$ g N kg^{-1} soil, where m_2 is a positive coefficient which needs to be

322 calibrated (Fig.2b). The inhibition effect of mineral N only occurs in case of

323 immobilization ($1/CN_{lit} < CUE_{opt}/CN_{SOM}$). The specific function $f(N_{min})$ can be

324 expressed as:

325
$$f(N_{min}) = \begin{cases} \min(1, m_2 \times N_{min}) \ , & \dfrac{CUE_{opt}}{CN_{SOM}} - \dfrac{1}{CN_{lit}} > 0 \\ 1 & , \quad \dfrac{CUE_{opt}}{CN_{SOM}} - \dfrac{1}{CN_{lit}} \le 0 \end{cases} \tag{5}$$

326   Existing studies have adopted approaches that differ from our definition to

327 explicitly represent the N inhibition effects on microbial processes (Manzoni and

328 Porporato, 2009; Bonan *et al.*, 2013; Fujita *et al.*, 2014; Averill and Waring, 2018). In

329 these previous studies, $f(N_{min})$ was assumed equal to the ratio between immobilized

330 mineral N and the N deficit for maintaining a stable C:N of decomposer biomass or

331 other receiver pools. Using the notation of Section 2, this definition of $f(N_{min})$ can be

332 expressed as:

333
$$f(N_{min}) = \begin{cases} \min\left(1, \dfrac{m_3 \times N_{min}}{U_0 \times \left(\frac{CUE_{opt}}{CN_{SOM}} - \frac{1}{CN_{lit}}\right)}\right) \ , & \dfrac{CUE_{opt}}{CN_{SOM}} - \dfrac{1}{CN_{lit}} > 0 \\ 1 & , \quad \dfrac{CUE_{opt}}{CN_{SOM}} - \dfrac{1}{CN_{lit}} \le 0 \end{cases} \tag{6}$$

334 where m_3 is a coefficient that needs to be optimized. U_0 (g C kg^{-1} soil day^{-1}) is the C

335 uptake rate (equivalent to the litter decomposition rate in absence of leaching) when

336 soil mineral N is fully adequate for litter decay (i.e. $f(N_{min}) = 1$), and can be calculated

337 from Eq. (7) as:

338
$$U_0 = C_{lit} \times k \times f(tem) \times f(water) \tag{7}$$

339 In this study, we also tested this formulation in the CENTURY-based model, in

340 addition to the Eq. (5) (see model M4 in Table A3).

341

342 2.5 Model parameterization and validation

343   To determine the respective impacts of including flexible CUE_d and N

344 availability constraining decay rates, we built five conceptual litter decay models

345 (Table 1). Model M0 corresponds to the default CENTURY parameterization of a

346 fixed CUE_d and no constraints of N availability on litter decay rates ($f(N_{min}) = 1$).

347 Model M1 accounts for flexibility in CUE from Eq. (2) and N constraints on decay

10

rates by Eq. (5). Model M2 has flexible CUE_d but no N constraints on decay rates ($f(N_{min})$= 1). Model M3 has N constraints on decay rates but a fixed CUE_d (Table 1). All of these four models are run at a daily time step. Finally, model M4 also accounts for flexibility in CUE and N constraints on decays (Table A3), but it uses Eq. (6) to represent the N constraints on decays rate rather than Eq. (2). Results from model M4 are presented in the main text, but only shown in the Appendix. This range of models allows identifying which mechanisms are at play during decomposition – flexible CUE_d only (M3), mineral N limitation only (M2), both mechanisms (M1, M4), or none (M0).

For calibrating model parameters and evaluation of their results, we collected data of laboratory litter incubation experiments from Recous *et al*. (1995) (5 experiments) and Guenet *et al*. (2010) (9 experiments, Table A2). The incubation experiments of Recous *et al*. (1995) and Guenet *et al*. (2010) continued 80 and 124 days, respectively. Recous *et al*. (1995) used corn residues (C:N = 130) and Guenet *et al*. (2010) used wheat straw (C:N = 44) in their incubation experiments. The C:N ratios of those corn residue and wheat straw span the range of litter C:N ratios among different ecosystems (Harmon *et al*., 2009; Brovkin *et al*., 2012; Manzoni *et al*., 2010). In the incubation experiments, plant litter was firstly cut into fine fragments before it was mixed with mineral soil. Soil temperature and moisture condition were kept constant during the experiment. Respired C from the incubated litter and SOC, as well as the soil mineral N concentrations were measured continuously across the incubation period. To distinguish the litter- and SOC-derived CO_2 flux, Guenet *et al*. (2010) used straw from wheat grown under ^{13}C labeled CO_2 and they are therefore able to track the CO_2 coming from litter and the CO_2 coming from soil. In the experiments by Recous *et al*. (1995), litter-derived CO_2 flux is calculated as the difference in CO_2 flux between the incubation samples with both soil and litter, and the control samples without added litter. More detailed information about the incubation experiments of Recous *et al*. (1995) and Guenet *et al*. (2010) can be found in Table A2.

The initial C storage and C:N ratios of litter and SOM pool, as well as soil

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temperature and moisture condition for decomposition in all of the five versions of the model (M0-M4) were set based on observations (Table A2). Plant litter was firstly separated into metabolic and structural litter pools based on its lignin to C ratio (LC_{lit}, dimensionless). The fraction of metabolic litter-C (f_m, 0-1, dimensionless) is calculated by:

$$f_m = f_{max} - m_4 \times LC_{lit} \tag{8}$$

where m_4 is a coefficient to be calibrated; f_{max}=0.85 is the maximum fraction of metabolic litter (i.e., the default value in CENTURY; Parton *et al.*, 1988). The fraction of structural litter-C is thus 1- f_m. The C:N ratios of both metabolic and structural pools are assumed to be equal to the C:N ratio of litter input.

In M1 and M3 models, the observed mineral N concentrations across the incubation period were used to calculate the daily N inhibition effect (Eq. (5)). The observed cumulative respired litter-C (g C kg^{-1} soil) measured in the incubation experiments was used to calibrate the model parameter values. Moreover, to quantify the simulated CO_2 flux derived from the litter, we also performed a set of control simulations with only SOM (initial litter pools were set to 0 g kg^{-1} soil) using the four model versions. The simulated litter-derived CO_2 flux is calculated as the difference in CO_2 flux between the simulation with both litter and SOM inputs and the simulation with only SOM input.

Parameter calibration was performed for each model with the shuffled complex evolution (SCE) algorithm developed by Duan *et al.*, (1993). The SCE algorithm relies on a synthesis of four concepts that have proved successful for global optimization: combination of probabilistic and deterministic approaches; clustering; systematic evolution of a complex of points spanning the space in the direction of global improvement and competitive evolution (Duan *et al.*, 1993). A more detailed description of this SCE optimization method can be found in Duan *et al.* (1993, 1994). In this study, the RMSE (root mean square error, Eq. (9)) between simulated and measured cumulative respired litter-C (%) on all observation days (Table A2) of each incubation experiment was used as the objective function, and the parameters minimizing RMSE between simulated and observed cumulative respired litter-C were

12

449 regarded as optimal parameter values.

450
$$RMSE = \sqrt{\left(\frac{\sum_{i=1}^{n}(Sim_i - Obs_i)^2}{n}\right)} \qquad (9)$$

451 where n is the number of observation days, Sim_i and Obs_i (%) are the simulated

452 and observed percent of cumulative litter-C flux on day i, respectively.

453   We used leave-one-out cross-validation (Kearns and Ron, 1997; Tramontana

454 *et al.*, 2016) to evaluate each of the four models (i.e. M0-M3), a cross validation

455 method used when data is scarce. The number of cross-validations corresponds to the

456 number of incubation experiments (14). Each time, one of the 14 incubation

457 experiments was left out as the validation sample, and the remaining 13 experiments

458 were used to train model parameters. In addition to RMSE, we also adopted the

459 Akaike Information Criterion (AIC, Bozdogan, 1987, Eq.(10)) to determine the

460 relative quality of the four version models on estimating cumulative respired litter-C.

461
$$AIC = n \times ln\left(\frac{\sum_{i=1}^{n}(Sim_i - Obs_i)^2}{n}\right) + 2n_p \qquad (10)$$

462 where n_p is the number of model parameters. The evaluation of AIC is important here

463 because depending on the model version, different numbers of parameters have to be

464 determined (Table 1), requiring us to weigh both model accuracy and robustness.

465   Note that the turnover times of SOM pools (active, slow and passive) used in

466 this study are obtained from the ORCHIDEE-MICT (v8.4.1, Guimberteau *et al.*,

467 2018). The turnover times of litter pools (metabolic and structural), as well as the

468 coefficient m_4 in Eq. (8) were optimized against the observed cumulative respired

469 litter-C from all of the 14 incubation experiments using the M0 and M1 models (Table

470 A3). A previous study has shown that litter decomposability is negatively correlated to

471 its physical size (for example, Tuomi *et al.*, 2011). Therefore, the turnover times of

472 the fine litter fragments used in the incubation experiments of Recous *et al.* (1995)

473 and Guenet *et al.* (2010) are expected to be shorter than the values set in

474 ORCHIDEE-MICT, which are representative of the turnover times of natural plant

475 residues. In addition, the mixing of soil and litter particle in the incubation experiment

476 likely enhances decomposition as spatial disconnection of decomposer and substrate,

477 which can occur under natural soil conditions (Barnes *et al.*, 2012; Hewins *et al.*,

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2013), is prevented. The calibrated turnover times of the metabolic and structural pools and the value of m_4 in Eq. (8) are 3.5 and 30 days and 0.5, respectively.

2.6 Impacts of litter stoichiometry and mineral N availability on SOM accumulation

We used the model M1, with flexible CUE_d and decomposition rate function of available N to study the impacts of litter stoichiometry (C:N ratio) and soil mineral N availability on the formation and accumulation of SOM. In total, 24 idealized simulation experiments with different values of litter C:N ratios and soil mineral N availabilities were conducted (Table A4). The assumed litter C:N ratios (CN_{lit}) of 10, 15, 30, 60, 120 and 200 span the variation among most natural substrates and soil amendments from organic matter input in agriculture (Harmon *et al.*, 2009; Brovkin *et al.*, 2012; Manzoni *et al.*, 2010). The assumed range of mineral N availability (N_{min}) of 0.001, 0.005, 0.01 and 0.05 g N kg^{-1} soil span the observed concentrations of soil mineral N in major terrestrial ecosystems (Metherall *et al.*, 1993).

In each simulation experiment, model M1was run for 5000 years to bring the litter and SOM pools in equilibrium with the prescribed litter input flux. The daily input rate of plant litter was set to 0.006 g C kg^{-1} soil day^{-1}, and the initial C stock of litter and SOM pools were all set to be 0 g C kg^{-1} soil. During the simulation, soil temperature and soil water content were assumed to be 25 ℃ and 60% of water holding capacity, respectively. We emphasized that our goal with this simplified scenario was to single out the effects of stoichiometric constraints, not to simulate the effects of a realistic climatic regime. Parameter values for M1 (with $m_1 = 0.54$, $n_1 = 0.50$ and $m_2 = 296.8$) used here were optimized based on all of the 14 incubation experiments from Recous *et al.* (1995) and Guenet *et al.* (2010) (see above). More detailed information about the specific settings of our simulation experiments can be found in Table A4.

3 Results

3.1 Evaluation of different models

Results of leave-one-out cross-validation suggest that model M1 provides more accurate prediction of cumulative respired litter-C than other models (Fig. 3). The differences between simulated and observed cumulative respired litter-C from M1 are mostly less than 6% for over 93% of the data (Fig. A1b). The average RMSE of predicted cumulative respired litter-C from M1 (3.0%) is lower than that of model M0 (4.1%). Models M2 and M3 have slightly lower RMSE values than M0 (3.7% and 3.8%, respectively) but perform worse than M1 (Fig.4). However, the average AIC of all the models are comparable, suggesting that models with more fitted parameters do not over-fit the observations (Fig. 4).

Model M1 captures the differences in respiration rates due to different C:N ratios of substrate and varying levels of mineral N availability across the 14 incubation experiments (Fig.5). While model M3 can reproduce the observed effect of soil mineral N availability on litter respirations rates (Fig.5d), it underestimates the cumulative respired CO_2 from low quality litter (CN_{lit} = 130) at high mineral N concentrations (> 0.04 g N kg^{-1} soil). Models M0 and M2 cannot represent the effects of soil mineral N on litter respiration rate (Figs. 5a, c), and their predictions are more biased from the observed values compared to M1. The Model M4, which uses the alternative formulation for N constraints on litter decay (Eq. (6)), reproduces the different respiration rates of substrates with contrasting C:N ratios and at different levels of mineral N availability (Fig. A2), but with a slightly higher average RMSE of cumulative respired litter-C than model M1.

The predicted CUE_d and the limitation effects of soil mineral N availability on litter decay rate ($f(N_{min})$ function in Eq. (5)) are different among the four tested models (Fig. A3). In models M0 and M3, which used a fixed CUE_d, the fitted values of CUE_d calculated with optimized parameters during the incubation period are about 0.57 and 0.54, respectively (Figs. A3a, d). In models M1 and M2, the CUE_d varies with the C:N ratios of plant litter, and is only slightly affected by soil mineral N concentrations (Figs. A3b, c). For very low quality litter with a C:N ratio of 130, the CUE_d in models M1 and M2 are 0.40 and 0.44, respectively, which are lower than for better quality litter with C:N ratio of 44 (approximately 0.55 and 0.56 in M1 and M2,

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respectively). Models M0 and M2 do not include the N inhibition effects on litter decay rate, thus the $f(N_{min})$ in these two models is always 1 (Figs. A3e, g). In M1 and M3, the N inhibition effect changes with both the litter C:N ratio and the mineral N availability (Figs. A3f, h).

CUE$_d$ from Eq. (2) calibrated with the data of the two incubation experiments, decreases with increasing CN$_{lit}$/CN$_{SOM}$ (Fig. 6). The average CUE$_d$ value is larger than the average of data compiled for microbial CUE of litter decomposition in terrestrial ecosystems by Manzoni *et al*. (2017). This is shown by the gray circles in Fig. 6. Our optimized values of CUE$_d$ for a given C:N ratio are more comparable with microbial CUE observed in incubations of soil mixed with litter (Gilmour and Gilmour, 1985; Dev êvre and Horw áth, 2000; Thiet *et al*.,2006), shown as black squares in Fig. 6.

3.2 The effect of litter quality *vs* quantity on equilibrium SOM stocks

Model M1 predicts that the size of the SOM pool at equilibrium is mainly determined by litter stoichiometry, with a minor effect of soil mineral N (Fig. 7). The lower C:N ratio of litter is, the higher equilibrium SOC stock. For litter with a specific C:N ratio, high soil mineral N concentration (e.g. above 0.05 g N kg^{-1} soil) generally produces a slightly larger equilibrium SOC stock than a low mineral N concentration (Fig. 7). Further analysis suggests that the SOC at equilibrium increases with decreasing litter C:N because the SOC pool is positively related to the CUE$_d$; however the limitation of soil mineral N on litter decomposition rate almost shows no impact on SOC (Fig. A4).

4 Discussion

We hypothesized that stoichiometric constraints (flexible CUE$_d$ or inhibition of decomposition under N limited conditions) played a role in shaping the trajectory of litter decomposition, with potential consequences on predicted SOC stocks. Our results suggest that flexible CUE$_d$ and inhibition effects of soil mineral N on litter

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decay rate improve prediction of litter decomposition when using a modified version of CENTURY model (denoted as M1). Evaluation of the model M1 using data from incubation experiments indicate that this modified model captures the effects of both variable litter quality (stoichiometry) and mineral N availability on respiration rates (Fig.5), without strongly inflating the complexity of CENTURY (Table 1). As the stoichiometric constraints are implemented in the generalizable and widely used structure of CENTURY and require only three parameters to be calibrated, they can also be easily implemented into land surface models for large spatial scale applications.

Accurately representing N control of microbial processes during litter decomposition has been suggested to be important for modeling the connection between the litter inputs, CUE_d, and soil C dynamics (Gerber *et al*., 2010; Manzoni *et al.*, 2012; Cotrufo *et al.*, 2013; Sinsabaugh *et al.*, 2013). In model M1, soil mineral N affects the litter-C flux via two mutually different pathways: (1) mineral N availability affects the litter decay rate and (2) flexible CUE_d determining the partition of decomposed C into SOC products and respired CO_2 (Fig. 1). Therefore, an increase in soil mineral N concentration enhances litter decay rates, which alone will increase the flux of litter-derived CO_2 (Eq. (5) and Fig. A5). However, as higher N concentration also results in a higher CUE_d (Eq. (2)), more C is transferred to SOC and less C is respired. In this way, SOC is predicted to accumulate with increasing mineral N availability when using model M1 (Fig. 7). In addition, the model M4, which is comparable to model M1 but uses an alternative formulation for N effects on the decomposition rate (Eq. (6)), performed slightly worse than model M1 (Fig. A2). Arguably, Eq. (6) represents the underlying mechanisms of N inhibition effects (Manzoni *et al.*, 2009; Bonan *et al.*, 2013; Fujita *et al.*, 2014; Averill and Waring, 2018) better than Eq. (5), and due to the minor differences in RMSE and AIC (Figure A2b) between these formulations it can serve as an alternative to M1.

Our results indicate that the observed diversity of responses of litter respiration rate to mineral N additions (Hobbie and Vitousek, 2000; Guenet *et al.*, 2010; Janssens *et al.*, 2010) is likely due to the combined effects of changes in litter

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decay rate and CUE_d (Fig. A5). Thus, N addition effects can differ among fertilization experiments if litter quality and background N availability vary. In addition to altering litter decay rate and CUE_d, mineral N addition can induce abiotic formation of compounds that resist microbial attack, inhibit oxidative enzymes involved in lignin degradation, stimulate microbial biomass production early in decomposition, or lead to the accumulation of microbial residues that are resistant to decay (Fog. 1988; Hobbie, 2015). All these effects might decrease litter respiration rate by inhibiting the decomposition process, but have not been considered in our current model.

This study provides insights on processes leading to increased SOM sequestration. Enhancing the efficiency at which plant residuals are transformed into stable SOM has been suggested as an effective strategy to sequester C in soil (Prescott, 2010; Cotrufo et al., 2013). Simulation results from our model suggest a positive linear relationship between equilibrium SOC stock and CUE of decomposed litter (Fig. A4), in line with the earlier findings with a similar model (for example Frey et al. 2013). In fact, with linear models such as CENTURY it can be shown that the steady state SOC scales linearly with CUE, different from nonlinear models predicting that higher CUE can trigger SOC loss (Allison et al., 2010). Our model goes beyond earlier attempts (Bonan et al., 2013; Fujita et al., 2014; Averill and Waring, 2018) by adapting the optimal metabolic regulation hypothesis of Manzoni et al. (2017) to link CUE, litter quality and SOM formation in a process-oriented way.

The importance of litter quality for SOM formation as found here is in line with recent experiments (Bahri et al., 2008; Rubino et al., 2010; Walela et al., 2014) and modeling studies (Grandy and Neff, 2008; Cotrufo et al., 2013). SOM is mainly formed though the partial decomposition of plant debris by microorganisms (Paul, 2007; Knicker, 2011; Cotrufo et al., 2013). The conceptual model developed by Cotrufo et al. (2013) suggested that although labile litter was decomposed faster than recalcitrant litter, a higher fraction of this labile litter-C would be incorporated into microbial biomass and subsequently incorporated into SOM pool (corresponding to a higher CUE_d). Therefore, labile litter inputs tend to form a larger SOM pool than the poor-quality (high C:N ratio) litter that is generally used by microbes at lower

efficiency. Our simulations of decomposition process of plant litter with different C:N ratios also suggest that litter of good quality (with low C:N ratio) can induce a larger SOM pool than the poor-quality litter (Fig. 7). CUE_d plays a more important role than the inhibition effect of low mineral N concentration in determining the size of the stable SOM pool (Fig. A4).

The predictions from Cotrufo *et al*. (2013) and this study contrasts with the conventional hypothesis whereby the poor-quality litter with low decay rate and small CUE_d are preferential to be accumulated in SOM (Berg and Mcclaugherty, 2008; Walela *et al.*, 2014).This view of SOM stabilization, however, seems to apply to N-limited systems with high C:N litter and where microbial remains are recalcitrant to decomposition (e.g., boreal forests) – in these systems SOC does accumulate despite its low quality (Kyaschenko *et al*. 2017). Moreover, one could argue that higher CUE_d implies larger microbial biomass, allowing faster decomposition (Allison *et al*., 2010). These feedbacks between microbial biomass and decomposition rate were not implemented in the current model, but could offer additional flexibility – again at the expense of more difficult model parameterization.

The CUE_d formulation from Eq. (2) with parameters calibrated from the two sets of incubation experiments might underestimate the impacts of litter quality on microbial CUE under natural conditions, in particular in case of SOM decomposition. In both incubation experiments, litter is firstly cut into fine fragments and then fully mixed with mineral soil (Recous *et al*.,1995; Guenet *et al*., 2010). Thus, the nutrient accessibility, air permeability and some other environmental factors (e.g. pH) of incubated litter are different from those of decaying litter in more natural, heterogeneous soil conditions. Those different decomposition conditions might be responsible for the differences observed in Fig. 6 between our CUE estimates and previously reported values. We speculate that more heterogeneous conditions reduce nutrient availability and thus might cause lower CUE. Similarly, CUE of surface litter decomposers may be lower than we estimated (Fig. 6), because litter not mixed with soil is probably subject to stronger nutrient limitation.

Further validation and development of our model are still necessary to

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decrease the model uncertainties. Soil mineral N which affects both litter decay rate and CUE of decayed litter is seldom monitored in litter incubation experiments (e.g. Walela *et al.*, 2014; Stewart *et al.*, 2015) and field litter decay experiments (e.g. Gholz *et al.*, 2000; Harmon *et al.*, 2009), with few exceptions (Recous *et al.*, 1995; Guenet *et al.*, 2010). An increasing number of land surface models (Wang *et al.*, 2010; Zaehle *et al.*, 2014; Goll *et al.*, 2017) have representations of the terrestrial N cycle. By incorporating our newly developed formulations of CUE_d and $f(N_{min})$ in these land surface models that simulate the dynamics of soil mineral N concentration, it will be possible to test and validate our developments with more extensive data from laboratory and field experiments. Moreover, similar to N, P has also been suggested as another important factor for litter decomposition and SOM formation (Güsewell and Verhoeven, 2006; Talkner *et al.*, 2009; Manzoni *et al.*, 2010; Prescott, 2010), especially in regions with highly weathered soil (Goll *et al.*, 2012, 2017; Yang *et al.*, 2014). So it might be necessary to include the effects of P on litter decay rate and CUE_d into our model for further decrease the simulation uncertainties.

5 Conclusions

By adapting the hypothesis of optimal microbial CUE proposed by Manzoni *et al.* (2017) for use in a CENTURY-based model and also introducing a N scaling function to represent the limits of mineral N availability on litter decay rate, we developed a simple but effective litter decomposition model that accounts for key stoichiometric constraints during decomposition. Validation using observation data obtained from laboratory incubation experiments indicated that our model could well predict the respiration rates of litter in different qualities at various levels of mineral N availability. Idealized simulations using our model revealed that the quality of litter inputs plays an important role in determining the soil C stock at equilibrium. High-quality litter (i.e. with low C:N ratio) tends to form a larger SOM pool as it can be more efficiently utilized by microorganisms than recalcitrant litter (e.g. high C:N ratio). Overall, the developed model captures the microbial mechanisms mediating

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1065 litter stoichiometry and soil mineral N effects on litter decomposition and SOM

1066 formation. Due to the simple and generalizable structure of our model, it can be

1067 incorporated into existing land surface models for further long-term and large spatial

1068 scale applications.

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Code and data availability

The CENTURY-based model used here is programmed in MATLAB language. The source code is available online (https://github.com/hchzhang/CENYUTY_CUE/tree/v1.0, DOI: 10.5281/zenodo.1307384). All the data used in this study can be obtained from published literatures. Specific references of these data can be found in section 2.5.

Competing interests

The authors declare that they have no conflict of interest.

Acknowledgements

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Table 1 Optimized parameter values for the five versions of the litter decomposition model used in this study. cue_{fit} is the optimized value of CUE, m_1 and n_1 are the coefficients in Eq. (3), m_2 is the coefficients in Eq. (5), and m_3 is the coefficient in Eq. (6). Values in brackets following each parameter are the means ($\pm$ standard deviations) of the fitted parameter values based on "leave-one-out" cross-validation (see Section 2.5 for more details).

Version	CUE	$f(N_{min})$	Parameters
M0	Fixed	1	cue_{fit} (0.57±0.004)
M1	Eqs.(2), (3)	Eq. (5)	m_1 (0.61±0.34), n_1 (0.53±0.21), m_2 (297.4±38.0)
M2	Eqs.(2), (3)	1	m_1 (0.11±0.01), n_1 (1.96±0.13)
M3	Fixed	Eq. (5)	cue_{fit} (0.54±0.01), m_2 (396.9±23.6)
M4	Eqs.(2), (3)	Eq. (6)	m_1 (0.13±0.07), n_1 (1.91±0.37), m_3 (0.58±0.12)

Figure 1 Schematic diagram of the C flows in the litter decay model used in this study. f_m is the fraction of metabolic compounds in plant litter. $D(C_{lit\text{-}met})$ and $D(C_{lit\text{-}str})$ are the decomposition rates (g C kg^{-1} day^{-1}) of metabolic or structural litter, respectively. LC_{lit} is the lignin:C ratio (on a mass basis) of plant litter; CN_{met}, CN_{str}, CN_{act}, and CN_{slow} are the C:N ratio of metabolic litter pool, structural litter pool, active SOM pool and slow SOM pool, respectively; N_{min} is the concentration of mineral N in solution (g N kg^{-1} soil); $f(N_{min})$ is a factor reducing litter decay rate when soil mineral N availability is limiting; T (℃) and SWC (%) are temperature and soil water content, respectively; CUE_d is C use efficiency of the transformation from litter to soil organic matter (SOM); $CUE_{max}=0.8$ is the maximum microbial CUE (dimensionless) when growth is limited by C from the organic substrate; f_{SA}, f_{SS} and f_{SR} are the fractions of decomposed structural litter-C that is transferred to active SOM pool, slow SOM pool and released to atmosphere in forms of CO_2, respectively. As in the algorithms in CENTURY model (Parton *et al.*, 1988), here $f_{SA}=CUE_{d\_SA}\times(1\text{-}f_{lig})$, $f_{SS}=CUE_{d\_SS}\times f_{lig}$, $f_{SR}=1\text{-}(f_{SA}+f_{SS})$, where f_{lig} is the lignin fraction (0–1, dimensionless) in the structural litter pool, and CUE_{d_SA} and CUE_{d_SS} are the CUE of C transformation from structural litter pool to active and slow SOM pool, respectively.

Figure 2. Schematic plot of (a) the optimal carbon use efficiency (CUE_{opt}) as a function of soil mineral nitrogen for different litter C:N ratios (from Eq. (2) in the main text with $m_1 = 0.3$, $n_1 = 1.0$) and (b) the N limitation function $f(N_{min})$ applied to litter decomposition rates (from Eq. (5) in the main text). CN_{lit} and CN_{SOM} are the C:N ratios of the litter and SOM pools, respectively. $CUE_{max} = 0.8$ is the maximum CUE under optimal nutrient condition (C limitation only). m_1 and n_1 are the parameters of Eq. (3) and m_2 is the parameter of Eq. (5).
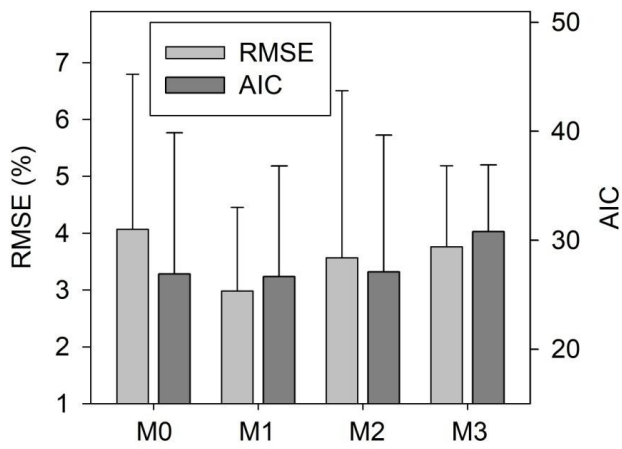
1468
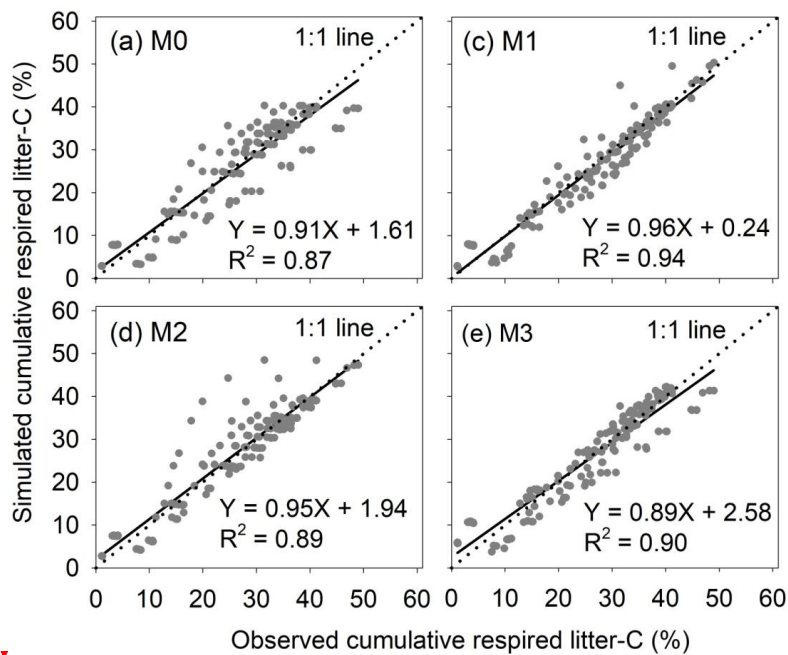
Figure 3 Comparison of the predicted cumulative respired litter-C to observed values
at different times during litter decomposition process. Each dot denotes an
observation of cumulative respired litter-C at a certain day. In total, there are 149

points. M0-M3 are the four versions of litter decay model tested in this study (Table
1).
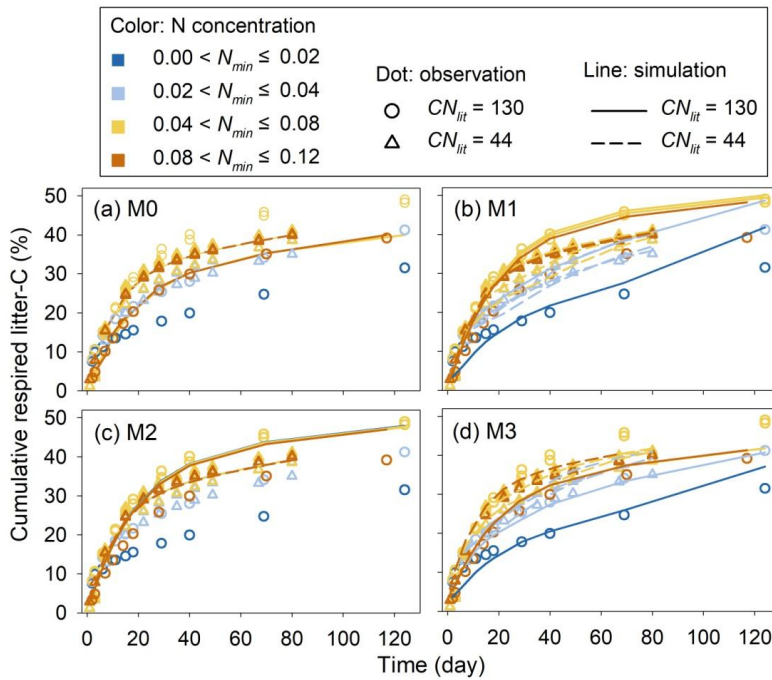
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Figure 4 The RMSE and AIC of the simulated cumulative respired litter-C from the four versions of litter decay model used in this study. Error bars denote the standard deviation of RMSE or AIC for different incubation experiments.M0-M3 denote the four models tested in this study (Table 1).
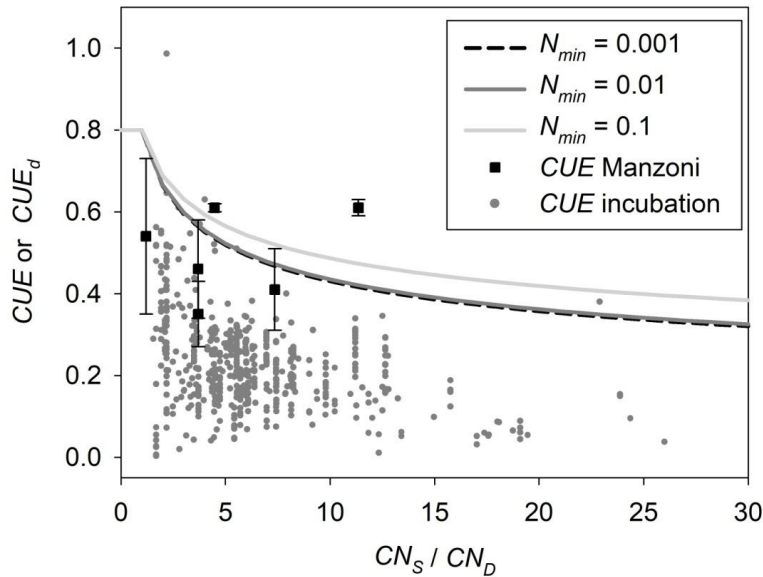
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Figure 5 Time series of the simulated (lines) and observed (dots) cumulative respired

litter-C (% of initial litter-C) at four different levels of soil mineral N availability (N_{min},

g N kg^{-1} soil). CN_{lit} is the C:N ratio of plant litter. M0-M3 denote the four models

tested in this study (Table 1). Here the simulation results of each model were

calculated with parameters optimized based on all of the 14 samples of incubation

experiments (Table A2).

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Figure 6 Comparison of CUE_d (lines) predicted by Eq. (2) with parameter values (m_2 = 0.54, n_1 = 0.50) calibrated based on the incubation experiments (Table A2) of Recous *et al*. (1995) and Guenet *et al*. (2010)to observed *CUE* of terrestrial microorganisms along a gradient of CN_S/CN_D. For observed CUE (dots), CN_D and CN_S are the C:N ratio of decomposers and their substrates, respectively. For simulated CUE (lines), CN_S and CN_D correspond to the C:N ratio of donor (litter pool) and acceptor (the active SOM pool of the CENTURY), respectively. Gray dots are the estimated microbial CUE of litter decomposition in natural terrestrial ecosystems from Manzoni *et al*. (2017). Black squares are the microbial CUE measured via labo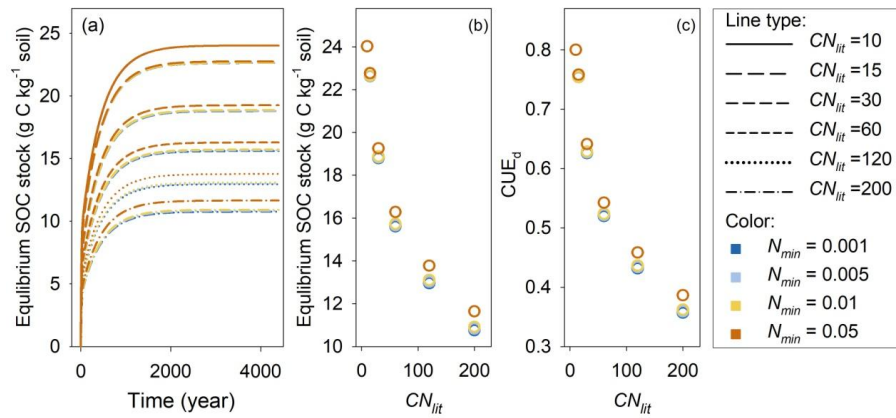ratory incubation experiments of Gilmour and Gilmour, (1985), Devêvre and Horwáth (2000) and Thiet *et al.* (2006). Error bars represent the standard deviations. N_{min} (g N kg^{-1} soil) is the concentration of soil mineral N.

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1512   **Figure7** (a) Accumulation of soil organic carbon (SOC) for constant substrates input

1513   (plant litter) with different C:N ratios (CN_{lit}) at different levels of soil mineral N

1514   concentrations (N_{min}, g N kg^{-1} soil), (b) Change trendsof equilibrium SOC stock and

1515   carbon use efficiency of decomposed litter (CUE_d) with increasing litter C:N ratio.

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Appendix:

Table A1 List of symbols used in this study; stoichiometric ratios are all expressed on a mass basis.

Symbol	Unit	Description
a	g N kg^{-1} soil	Exponent in Eq. (2)
AIC	dimensionless	The Akaike Information Criterion(Eq. (10))
CN_{act}	dimensionless	C to N ratio of active soil organic matter pool
CN_D	dimensionless	C to N ratio of decomposer (Eq. (1))
CN_{met}	dimensionless	C to N ratio of metabolic litter pool
CN_{slow}	dimensionless	C to N ratio of slow soil organic matter pool
CN_{str}	dimensionless	C to N ratio of structural litter pool
CN_S	dimensionless	C to N ratio of substrate (Eq. (1))
C_{lit}	g C kg^{-1} soil	C stock of litter pool(Eq. (4))
CN_{lit}	dimensionless	C to N ratio of litter pool (metabolic or structural, Eq. (2))
CN_{SOM}	dimensionless	C to N ratio of soil organic matter pool
CUE	dimensionless	Microbial carbon use efficiency
CUE_d	dimensionless	Carbon use efficiency of decomposition (C incorporated in SOC over litter C decomposed)
CUE_{fit}	dimensionless	Optimized value of fixed CUE in model M0 and M4
CUE_{max}	dimensionless	Maximum CUE$_d$ (Eqs. (1) and (2))
CUE_{opt}	dimensionless	Optimal CUE$_d$ (Eq. (1))
CUE_{d_SA}	dimensionless	CUE of the transformation from structural litter to active SOM pool
CUE_{d_SS}	dimensionless	CUE of the transformation from structural litter to slow SOM pool
$D(C_{lit\text{-}met})$	g C kg^{-1} soil day^{-1}	Decomposition rate of metabolic litter
$D(C_{lit\text{-}str})$	g C kg^{-1} soil day^{-1}	Decomposition rate of structural litter
$f(N_{min})$	dimensionless	Limit factor of soil mineral N on litter decomposition (Eqs. (4) and (5))
$f(tem)$	dimensionless	Limit factor of soil temperature on litter decomposition (Eq. (4))
$f(water)$	dimensionless	Limit factor of soil water content on litter decomposition (Eq. (4))
f_m	dimensionless	Fraction of metabolic plant litter
f_{SA}	dimensionless	Fractions of decomposed structural litter-C that is transferred to active SOM pool
f_{SR}	dimensionless	Fractions of decomposed structural litter-C that is released tp atmosphere
f_{SS}	dimensionless	Fractions of decomposed structural litter-C that is transferred to slow SOM pool
I_N	g kg^{-1} soil	Maximum mineral N immobilization rate (Eq. (1))
k	day^{-1}	potential maximum turnover rate (Eqs. (4) and (7))
LC_{lit}	dimensionless	Lignin to C ratio of litter input
m_1	kg g^{-1} N	Coefficients in Eq. (3)
n_1	g N kg^{-1} soil	Coefficients in Eq. (3)
m_2	day^{-1}	Coefficients in Eq. (5)

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m_3	kg g^{-1} N	Coefficients in Eq. (6)
N_{min}	g N kg^{-1} soil	Soil mineral N concentration (Eq. (5))
RMSE	%	Root mean square error (Eq. (9))
SOC	g C kg^{-1} soil	Soil organic carbon
SOM	g C kg^{-1} soil	Soil organic matter
U_0	g C kg^{-1} soil day^{-1}	C uptake rate when soil mineral N is fully adequate for litter decay (Eqs. (1) and (8))

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Table A2 Information about the 14 samples of laboratory incubation experiment used in this study. CN_{lit} and LC_{lit} are the C to N ratio and lignin to C ratio of plant litter, respectively. CN_{SOM} is the C to N ratio of SOM pool. N_{min} is the concentration of soil mineral N (NO_3^- -N + NH_4^+ -N). For the incubation experiments of Guenet *et al.* (2010), cumulative respired litter-C was measured on days 1, 3, 7, 15, 22, 28, 35, 42, 49, 67 and 80, and N_{min} was measured on days 3, 7, 17, 28 and 80. For the incubation experiments of Recous *et al.* (1995), both cumulative respired litter-C and N_{min} were mostly measured on days 2, 3, 6, 11, 15, 18, 29, 40, 69 and 124.

Sample	CN_{lit}	LC_{lit}	CN_{SOM}	Initial N_{min} (g N kg^{-1} soil)	Duration (day)	Temperature (℃)	Soil moisture (%, in volume)	Litter type	Reference
1	44	0.26	11	0.035	80	20	50	Crop (wheat)	
2	44	0.26	11	0.051	80	20	50	Crop (wheat)	
3	44	0.26	11	0.055	80	20	50	Crop (wheat)	
4	44	0.26	11	0.033	80	20	50	Crop (wheat)	Guenet *et al.*, 2010
5	44	0.26	11	0.049	80	20	50	Crop (wheat)	
6	44	0.26	11	0.067	80	20	50	Crop (wheat)	
7	44	0.26	11	0.033	80	20	50	Crop (wheat)	
8	44	0.26	11	0.048	80	20	50	Crop (wheat)	
9	44	0.26	11	0.079	80	20	50	Crop (wheat)	
10	130	0.23	9	0.010	124	15	42	Crop (Corn)	
11	130	0.23	9	0.030	124	15	42	Crop (Corn)	Recous *et al.*, 1995
12	130	0.23	9	0.060	124	15	42	Crop (Corn)	
13	130	0.23	9	0.080	124	15	42	Crop (Corn)	
14	130	0.23	9	0.100	124	15	42	Crop (Corn)	

1532 **Table A3** List of parameters calibrated for two versions of the litter decomposition

1533 model (M0, M1): k_{litm} and k_{lits} are respectively the turnover rates of metabolic and

1534 structural litter pools, m_4 is the coefficient in Eq. (8), cue_{fit} is the optimized value of

1535 CUE, m_1 and n_1 are the coefficients in Eq. (3), and m_2 is the coefficients in Eq. (5).

Version	CUE	$f(N_{min})$	Parameters
M0	Fixed	1	$cue_{fit}, k_{litm}, k_{lits}, m_4$
M1	Eqs. (2), (3)	Eq. (5)	$m_1, n_1, m_2, k_{litm}, k_{lits}, m_4$

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1547 **Table A4** Specific setting of litter and SOM properties, and soil conditions in the 16

1548 idealized simulations for exploring the impacts of litter stoichiometry (i.e. C:N ratio)

1549 and soil mineral N on SOC accumulation. CN_{lit} and LC_{lit} are the C to N ratio and

1550 lignin to C ratio of plant litter, respectively. Lit_{inp} (g C kg^{-1} soil day^{-1}) is the daily

1551 input rate of plant litter. CN_{SOM} is the C to N ratio of SOM pool. N_{min} (g N kg^{-1} soil)

1552 is the concentration of soil mineral N (NO_3^- -N + NH_4^+ -N). *Tem* (°C) and *SWC* (%)

1553 are the temperature and soil water content, respectively.

Experiment	CN_{lit}	LC_{lit}	Lit_{inp}	CN_{SOM}	N_{min}	*Tem*	*SWC*
1	15	0.2	0.006	12	0.001	25	60
2	30	0.2	0.006	12	0.005	25	60
3	60	0.2	0.006	12	0.01	25	60
4	120	0.2	0.006	12	0.05	25	60
5	15	0.2	0.006	12	0.001	25	60
6	30	0.2	0.006	12	0.005	25	60
7	60	0.2	0.006	12	0.01	25	60
8	120	0.2	0.006	12	0.05	25	60
9	15	0.2	0.006	12	0.001	25	60
10	30	0.2	0.006	12	0.005	25	60
11	60	0.2	0.006	12	0.01	25	60
12	120	0.2	0.006	12	0.05	25	60
13	15	0.2	0.006	12	0.001	25	60
14	30	0.2	0.006	12	0.005	25	60
15	60	0.2	0.006	12	0.01	25	60
16	120	0.2	0.006	12	0.05	25	60

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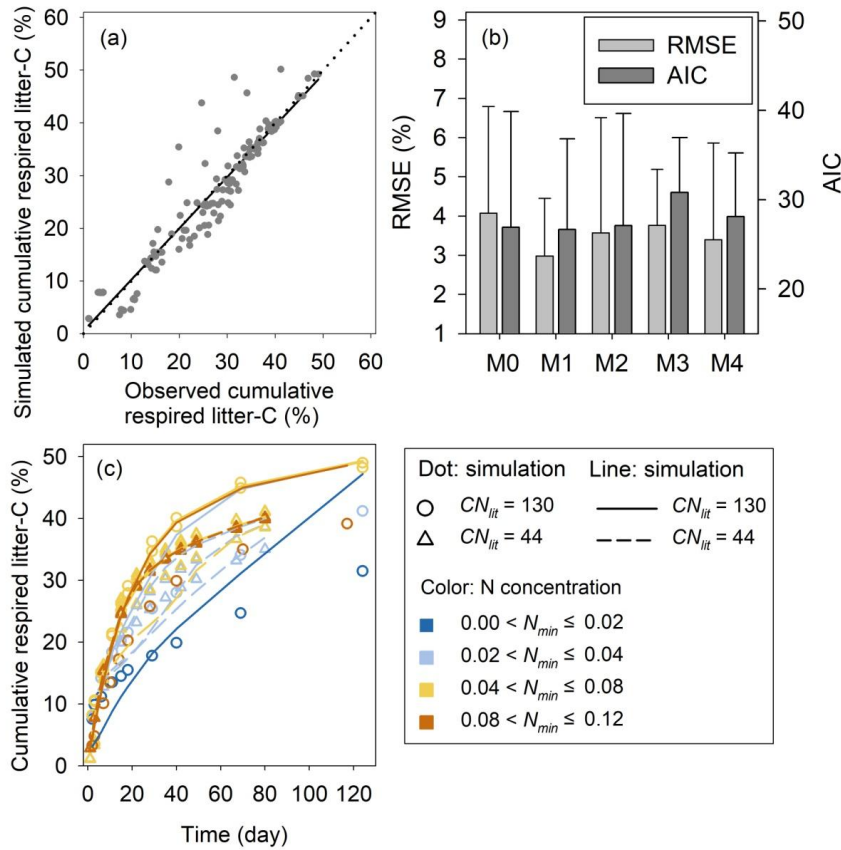删除的内容: A3
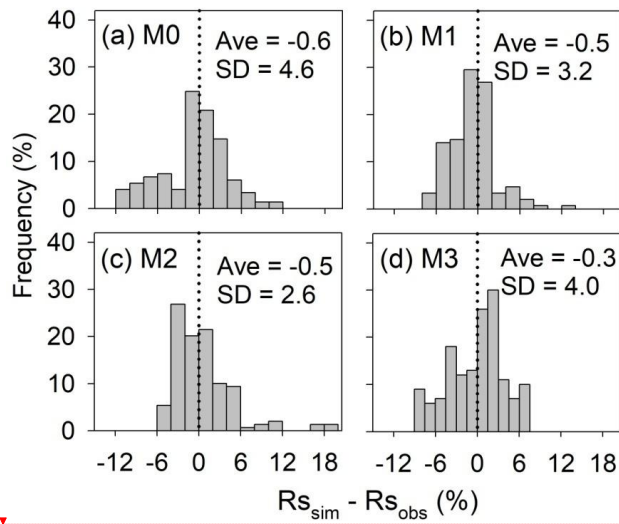
Figure A1 Distribution of the difference between the predicted cumulative respired litter-C (Rs_{sim}, %) and the observed values (Rs_{obs}, %) for all experiments and points in time. SD is standard deviation of the biases. M0-M3 denote the four models tested in this study (Table 1).

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Figure A2 Comparison between simulated cumulative respired litter-C with $f(N_{min})$ (inhibition effect of soil mineral N on litter decomposition) calculated by Eq. (6) and the observed results from incubation experiments. In figure (c), M0-M4 denote the five versions of litter decay model in Table 1. M4 denote the model which used Eq. (2) to calculate the dynamic CUE and Eq. (6) to calculate $f(N_{min})$.
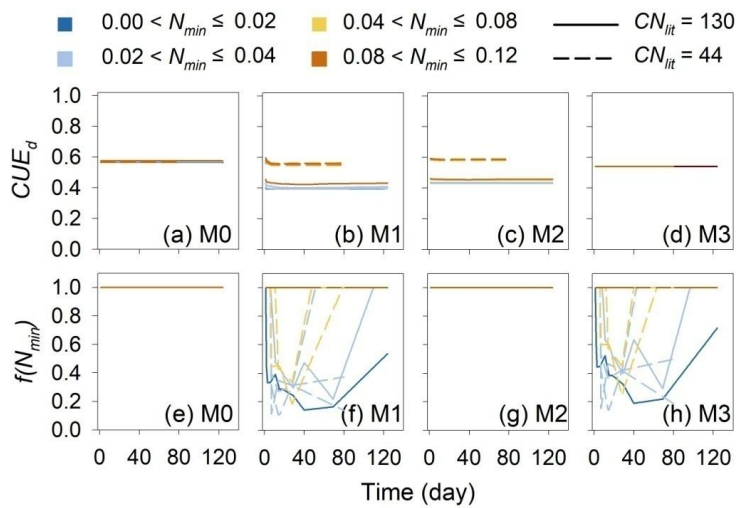
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Figure A3 Dynamic of the simulated carbon use efficiency (*CUE*) and *f*(N_{min}) during

the incubation experiments (Table A4). CN_{lit} is the C:N ratio of incubated litter, and

N_{min} is the initial soil mineral N concentration (g N kg^{-1} soil). M0-M3 are the four

models in Table 1. Here the simulation results of each model were calculated with

parameters optimized based on all of the 14 samples of incubation experiments
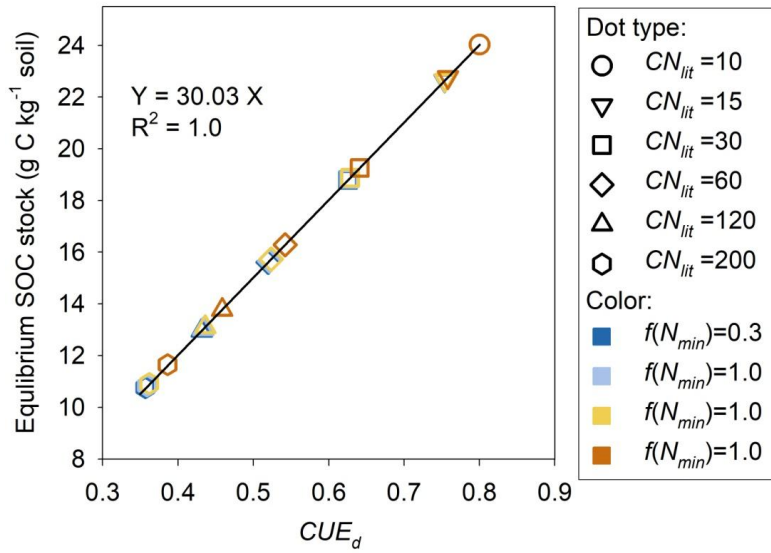
(Table A2).
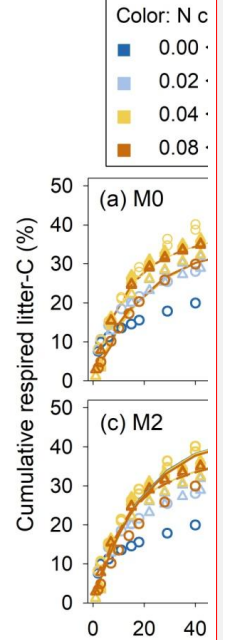
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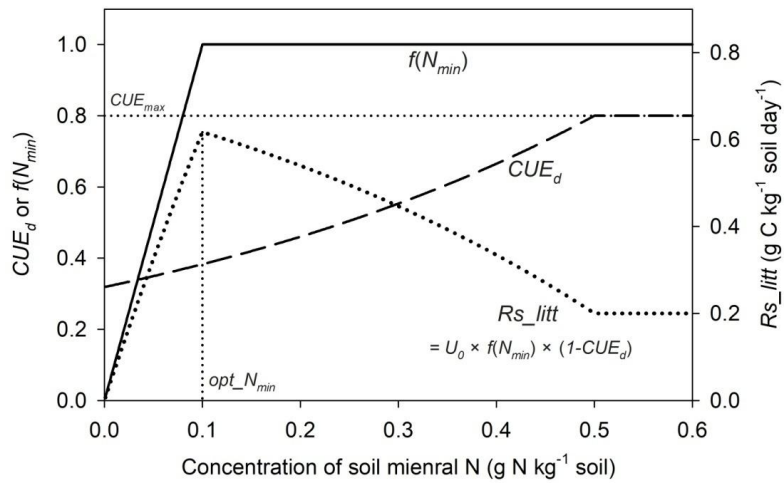
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Figure A4 Relationship between C stock of the potentially equilibrated SOM pool

and the carbon use efficiency of decomposed metabolic litter (CUE_d) at the dynamic

equilibrium stage. $f(N_{min})$ denote the inhibition factor (0–1) of soil mineral N on litter

decomposition.

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Figure A5 Schematic plot for change trends of $f(N_{min})$ (inhibition effect of mineral N,

Eq. (6)), CUE_d (carbon use efficiency of decomposed litter, Eqs. (2), (3)) and Rs_litt

(litter respiration rate) with increasing concentration of soil mineral N. CUE_{max} (=

0.8) is the maximum CUE set in this study. opt_N_{min} denotes the concentration of

soil mineral N at which litter respiration is maximized. U_0 is the potential

decomposition rate when mineral N is fully adequate for litter decay.

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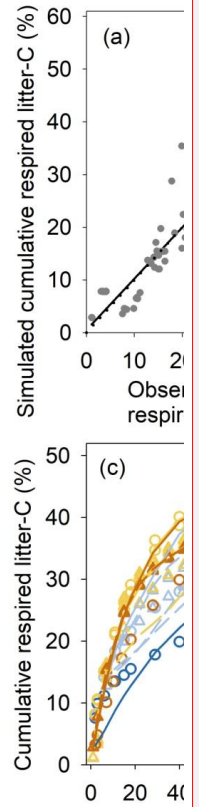
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Figure A5 Comparison between simulated cumulative respired litter-C with $f(N_{min})$ (inhibition effect of soil mineral N on litter decomposition) calculated by Eq. 9 and the observed results from incubation experiments. In figure (c), M0-M3 denote the four versions of litter decay model in Table 1. M4 denote the model which used Eq. 2 to calculate the dynamic CUE and used Eq. 6 to calculate $f(N_{min})$.

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