Dr Maria Val Martin
UKRI Future Leaders Fellow
LC3M, University of Sheffield
E-mail: m.valmartin@sheffield.ac.uk
https:/mariavalmartin.wordpress.com/

The University Of Sheffield.

Sheffield, August 21, 2023

RE: Improving nitrogen cycling in a land surface model (CLM5) to quantify soil N_2O, NO and NH_3 emissions from enhanced rock weathering with croplands

Dear Chris Folberth,

Thank you again for managing the reviews of our manuscript. We have made several changes in the manuscript in response to Reviewer 3' comments.

Following with your suggestion, we have also clarified the new capabilities introduced in CLM5 within the Abstract. Specifically, we now outline the model structure, including the direct implementation of new routines within the CLM5 nitrogen cycle, along with the offline coupling of soil pH from the ERW model. This change aims to provide readers with a clearer understanding of our model's advancements from the start.

We acknowledge your recommendation to include a brief critical reflection in the conclusions. We would like to highlight that, in response to one of the comments from Reviewer 1, we had already integrated such a reflection. In the Conclusions section (Lines 526-532), we explicitly state that ongoing efforts are needed to improve the nitrogen cycling representation in CLM5 and to further refine the implementation of enhanced rock weathering, especially as more field trial data become available.

We are thankful to the three reviewers for their valuable comments and feedback. Their insights have significantly contributed to the improvement of our manuscript. In accordance with the structure recommended by GMD, we have provided a detailed response to each reviewer's comments.

We believe that these revisions effectively address the concerns raised by the reviewers and you. All co-authors agree and fully support the submission of this revised version of our work.

Thank you once again for your time and consideration.

Sincerely,

Maria

Dr Maria Val Martin

REVIEWER 3

Maria Val Martin et al. present their study on enabling the CLM5 to quantify the changes in soil N2o, NO and NH3 emissions due to enhanced rock weathering in croplands. The topic is very interesting and important. Nonetheless, I still have a few concerns on this paper:

We are glad the reviewer finds this paper interesting and important. We have responded to each comment sequentially with italicised text showing the reviewer's comments and plain text showing our response. New text added to the manuscript is coloured blue, and any text removed from the manuscript is struck through. The locations of changes are stated.

1) I was very exciting when I saw the title of this paper, as I have thought that the authors have merged the effects of enhanced rock weathering on soil pH and N cycle in CLM5. But I am disappointed when I know that the authors actually did not couple the rock weathering process into the CLM5. The changes in soil pH caused by rock weathering have been simulated offline by the ERW model. Then the CLM5 used the simulated soil pH from the ERW model as a forcing data. As shown in previous studies (e.g. Beerling et al., 2020; Kantzas et al., 2022), dynamics of soil N cycle can also affect rock weathering. Without a fully-coupled model, it might be hard for the revised CLM5 to correctly capture the interactions between soil N cycle, soil pH and rock weathering. Implementing rock weathering in CLM5 should be the most important contribution of this study. It is a pity that the authors have not done it.

We note the reviewer's desire for a paper in which there is a full coupling of the ERW model within CLM5. However, the first step has to be improving and evaluating the CLM5's nitrogen cycle response to ERW and this is the purpose of out paper – as clearly specified in the title. Our paper exploits the significant progress that has been made in understanding key drivers of soil N- cycle responses to ERW effects including, for example, soil pH from recent ERW field trials and mesocosm experiments (Blanc-Betes et al., 2020, Chiaravalloti et al., 2023). Our approach focuses on leveraging these advancements for model development, feeding into impacts on agroecosystem N cycle mechanisms and subsequent effects on soil nitrogen emissions. This represents a major advancement in a field that remains at its early stages, despite its important potential in climate change mitigation.

Thus, our intention is to equip the CLM5 land model with new capabilities that allow reproducing what to date is thought to be a key driver of N biogeochemistry in soils under this promising climate stabilization strategy.

Importantly, our approach captures interactions between the soil N cycle, soil pH, and ERW as comprehensively as possible, with an asynchronous coupling of the ERW and CLM5 models, as described in Kantzas et al., (2022). Initially, we run the ERW model with CLM5-derived inputs (e.g., climate, water infiltration, soil pH, N changes, CO_2 respiration, etc.). Subsequently, we use the ERW-derived soil pH changes to drive CLM5 and estimate changes in soil N emissions. Thus, the dynamics of soil N cycle that can affect ERW are capture by depth in the soil profile and by time for any individual grid cell.

To avoid misleading the readers, we have taken steps to clarify this aspect. We have improved the abstract and introduced a new section (2.2.4), as suggested below, to elaborate on how the coupling of soil pH is achieved in our model.

Abstract

> We base the new parameterizations on datasets derived from soil pH responses of N_2O, NO and NH_3 of ERW field trial and mesocosm experiments with crushed basalt. These new capabilities involve the direct implementation of routines within the CLM5 N-cycle

framework, along with asynchronous coupling of soil pH changes estimated through an ERW model.

Lines 220-230

2.2.4 Weathering

To simulate the impact of basalt addition on soil N_2O, NO, and NH_3 fluxes, we introduced a weathering option into CLM5. This approach involves incorporating annual or monthly changes in soil pH estimated by an ERW model (Beerling et al., 2020; Kantzas et al., 2022) into the CLM5 N cycle. The coupling of soil pH in CLM5 and the ERW model occurs asynchronously (Kantzas et al., 2022). In the first phase, the ERW model dynamically calculates soil pH using alkalinity mass and flux balances with an adaptive time-step controlled by mineral dissolution rates. The alkalinity balance accounts for net acidity input during crop growth for removed biomass cations, and secondary mineral precipitation of calcite. Additionally, the N cycle's influence on soil acidity is considered. For that, each nitrogen transformation (e.g., nitrification, denitrification, volatilization) is associated with hydrogen ions production or consumption, leading to stoichiometric acidity fluxes to each nitrogen flux within the ERW model. These calculations begin with initial soil pH values and nitrogen fluxes provided by CLM5 to the ERW model, run individually at each grid cell. Subsequently, in the second phase, spatially distributed changes in soil pH (i.e., delta pH) estimated by the ERW model are integrated into CLM5. This process enables CLM5 to adjust the initial soil pH values accordingly. Detailed descriptions of the soil pH calculations are provided in Beerling et al., (2020) and Kantzas et al., (2022).

2) Lines 242-250 described how the effects of rock weathering on soil N cycle is simulated in this study. Thus, I suggest to move these lines to the section 2.2 (maybe add a sub-section 2.2.4). In addition, I would suggest the authors to add some text to explain how the effect of weathering on soil pH is simulated in the ERW model.

Modified as suggested. We added now a section 2.2.4 (see above) and updated section 2.3 as follows:

Lines 260-266

To include ~~model~~ the effect of ERW on the N_2O, NO and NH_3 fluxes from soil, ~~we developed a weathering option for CLM5, in which annual or monthly changes in soil pH estimated by an ERW model offline (Beerling et al., 2020; Kantzas et al., 2022) are read within the CLM5 N cycle. Specifically, CLM5 acquires spatially distributed delta pH values, and adjusts the initial soil pH accordingly.~~ we considered the soil pH changes as well as application locations across five key regions with high potential for CDR with ERW (North America, Brazil, Europe, India, and China) required to remove 2Gt CO_2 per year (Beerling et al., 2020). Thus, in the "Control" Run soil pH is kept constant to the nominal values provided by Harmonized World Soil Database, whereas in the "ERW" Run is modified following the ERW model projection. To test the new scheme at a global scale, we used changes in annual soil pH (Fig. 2 and Fig. S5 in SM); dynamic changes of soil pH in monthly timesteps were tested in a regional study for the UK (Kantzas et al., 2022).

3) I agree with the previous two authors that the validation of the simulation results is too weak in this study. The simulated N2O fluxes have at least been evaluated against some site-level observations. But for NO and NH3, there is no validation. As indicated by referee #1, the model seems not ready for a global application.

As stated in our earlier response to Reviewers 1 and 2, in this study we have evaluated our model against field observations for N_2O from two crops with distinct physiology (e.g., high vs low nutrient requirements) and management practices (e.g., N fertilizer loads) These crops also experienced different levels of soil pH changes, enabling us to assess the model's performance across a range of conditions and crop requirements. Furthermore, we compared our model results against multiple global emission inventories to provide additional context and validation and showed that CLM5 successfully provides a reasonable representation of the magnitude and seasonality of direct agriculture nitrogen emissions in major cropland regions.

The agriculture NH_3 emission parameterization used in our study was originally implemented by Fung et al. (2022) and has undergone thorough evaluation, including a global sensitivity analysis to assess the impact of soil pH on soil NH_3 fluxes. We also note that the new implemented regulating ERW pH-NH_3 function includes new observations from a mesocosms experiment with basalt application within a greenhouse setting, a dataset shared with us for the specific purpose of this work by Chiaravalloti and colleagues (Chiaravalloti et al., 2023). Similarly, the soil NO parameterization we employed is a well-established approach from Parton et al. (2001), which has been implemented in various land vegetation models (e.g., CLM5, PSL, and ORCHIDEE), and is widely used across the earth system modelling community. These established parameterizations provide a solid foundation for our modelling framework.

Furthermore, we respectfully disagree with the statement that the model is not ready for a global application. As suggested by Reviewers 1 and 2 and the Editor, we have included a critical reflection in Conclusions, in which we acknowledge that our study is a first step towards improving the nitrogen cycling-ERW representation in CLM5.

Throughout the manuscript, we have taken care to emphasize that our implementations represent an initial attempt at parameterizing ERW in a land model, and that ongoing efforts are necessary to refine the ERW scheme. We have now clarified this further in the Abstract as well.

Abstract

> While further developments are still required in our implementations when additional ERW data become available, our improved N-cycle scheme within CLM5 has utility for investigating the potential of ERW point-source and regional effects of soil N_2O, NO and NH_3 fluxes in response to current and future climates.

4) In Table 2&3, Fig. 4&6, I would suggest to add the result of the default CLM5 which does not consider the rock weathering. This will help the readers to know if the revised model performs better than the default model.

We appreciate the reviewer's keen interest in comparing the results of the default CLM5 with the revised model. We want to clarify that the primary objective of our study was not to improve the estimates of soil N_2O fluxes within CLM5. Instead, our focus was in implementing parameterizations that enable the representation of changes in in soil N_2O emissions driven by soil pH alterations, alongside the estimation of soil NO and NH_3 fluxes, which were not accounted for in the default version. As suggested, we have included the soil N_2O fluxes derived from the default model only in Tables 2 and 3 for simplicity and clarity for the Figures and added a discussion in the manuscript accordingly. The discussion reads now as:

Lines 400-404 Section 3.2

For N_2O, CLM5 estimates global direct agriculture emissions of 3.1 Tg N_2O-N yr^{-1}, which is in line with previous annual estimates for agriculture sources (1.7–5.8 Tg N yr^{-1}; e.g., Del Grosso et al., 2006; Syakila and Kroeze, 2011; Saikawa et al., 2014) and the IPCC 2021 reported values for 2007–2016 (3.8 Tg N yr^{-1}) (Canadell et al., 2021). Our updates in the CLM5 N-cycle did not significantly alter the global soil N_2O flux in agriculture systems compared to the default CLM5 version (4.2 Tg N_2O-N yr^{-1}). In addition, our estimate is similar [….]

Lines 442-452 Section 3.2

In CLM5, major crop N_2O emitters are Europe (0.68 Tg N yr^{-1}), China (0.63 Tg N yr^{-1}) and North America (0.59 Tg N yr^{-1}), each with about 19–22% of global emissions. Our modifications to the CLM5 N-cycle did not result in significant regional-scale changes in agriculture soil N_2O compared to the default version, although led to lower N_2O emissions over India (0.18 vs. 0.36 Tg N yr^{-1}).

5) Why does the spin-up method for site-level simulation differ from that used for global simulation?

The spin-up methodology employed for both site-level and global simulations are the same, which involved an initial accelerated spin-up phase, followed by a historical simulation. The global simulation's accelerated spin-up duration was however extended to 1200 years, contrasting the single-site simulation's 600-year period. This longer time was necessary due to the Arctic region's substantial soil organic matter carbon, requiring an extended duration to achieve equilibrium. We have revised the description in the manuscript to enhance the clarity of the spin-up method.

Lines 227-229

The meteorological forcings were from the Global Soil Wetness Project (GSWP3 version 1; http://hydro.iis.u-tokyo.ac.jp/GSWP3/), with forcing data available from 1901 to 2014 and cycled from 1901 to 1920 for years prior to 1901.

Lines 240-245

The accelerated decomposition spin-up was for about 1200 years as the total soil organic matter carbon in the Arctic regions requires a longer time frame to reach equilibrium; we considered the model fully spun-up when […]

As in the single-point simulations, the present-day spin-up was based on a historical simulation 1850–2014, using historical N and aerosol deposition, atmospheric CO_2 forcing, land use change and meteorological forcings from GSWP3 (Lawrence et al., 2019). ~~The meteorological forcings were from the Global Soil Wetness Project (GSWP3 version 1; http://hydro.iis.u-tokyo.ac.jp/GSWP3/), with forcing data available from 1901 to 2014 and cycled from 1901 to 1920 for years prior to 1901.~~

REVIEWER 1 or 2

Maria Val Martin et al. has addressed all my major comments. Lastly, I would like to suggest adding a few sentence to clarify the offline ERW model in terms of how to estimate the monthly change of pH for CLM, around line 252. (This is a critical capability that has been added into CLM).

Thanks a lot for your supportive comment. As also suggested by Reviewer 3, we have added a new section 2.2.4 on Weathering that explains how CLM5 treats the soil pH from the ERW model and clarifies how the monthly changes in soil pH are estimated.