REVIEWER 1

Maria Val Martin et al., presented an interesting study that developed new capability for CLM5 N cycle and used this new model to investigate the impact of ERW-induced pH changes on agriculture soil N20/NO/NH4 emissions. This is an exciting and timely work that greatly advances existing modeling tool and will benefit the research community a lot. Below are my comments and suggestions.

We are glad the reviewer finds this paper exciting and timely. 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.

The model development description is very clear; however, the model simulation plan needs to be improved. Important information is missing, and the choice of forcing data is potentially problematic. For example, section 2.3, which forcings data are used for spin-up simulation? How many years of simulation were conducted for accelerated spinup versus regular spinup? What are the criteria of soil N variables for a fully-spunup condition? Why use GSWP3 forcings for 1850-2014 simulation but NLDAS forcings for 2015-2019? When transition from 2014 to 2015, the difference between NLDAS and GSWP3 forcings will result in sharp change of vegetation and soil dynamics. For global simulations, again, how many years of accelerated spinup versus regular spinup were performend? How to determine if global land has reached a fully-spunup condition.

We thank the reviewer for pointing out that our model simulation process needed further clarification. In terms of the single-point spin-up, we followed the recommended guidelines outlined in the CLM5 User Guide. Specifically, we conducted an accelerated decomposition spin-up, spanning 200 years, starting from a cold-start condition. Additionally, we extended the spin-up by approximately 400 years through the regular decomposition simulation. Throughout the spin-up process, we thoroughly analyzed various state variables within the model, such as total ecosystem carbon, total soil organic matter carbon, total water storage, soil N_2O, LAI, and more. We observed that the soil nitrogen species reached equilibrium considerably sooner (15-20 years) compared to the carbon species (>400 years).

In the case of global simulations, we followed similar guidelines but with longer spin-up times (400 years followed by an additional 800 years). To ensure that the model reached equilibrium, we monitored the disequilibrium of the land surface, aiming for a threshold of less than 3% as determined by the total ecosystem carbon. It is important to note that the total soil organic matter carbon exhibited a longer time frame to reach equilibrium, particularly in Arctic regions, and a criterion of less than 3% disequilibrium is considered acceptable.

We have expanded the description of the model spin-up process for both single-point and global simulations in Section 2.3 of the manuscript, accordingly.

Section 2.3 Line 219-223

> We performed single-point simulations at the Energy Farm field site (University of Illinois, U.S.) to examine the model sensitivity to basalt applications in maize and soybean crops and soil and climate conditions. We spun-up the model for about 600 years, so that all the state variables in the model, especially total ecosystem soil carbon and soil N_2O reached equilibrium. Then, the same initial condition was used for both the soybean and the corn single-point present-day spin-up simulations because a uniform soil condition was achieved for both crop systems.

Section 2.3 Lines 234-236

> We first spun-up CLM5 with the new implementations to steady state in 1850 using an accelerated decomposition procedure and fixed pre-industrial CO_2, land use, and atmospheric N deposition (Lawrence et al., 2019). The accelerated decomposition spin-up was for about 1200 years and we considered the model fully spun-up when the land surface had more than 97% of the total ecosystem carbon in equilibrium. After the historical spin-up, we initialized CLM5 simulations for 2000 using fully spun-up conditions.

The reviewer made a valid point highlighting the potential influence of using different atmospheric forcings on vegetation and soil dynamics. The decision to use both GSWP3 and NLDAS atmospheric forcings stemmed from data availability limitations. Our objective was to simulate soil N_2O emissions at the Energy Farm, specifically up to 2019, in order to compare them with available observations from 2016-2019. However, the GSWP3 data are only provided up to 2014, while the NLDAS database covers the period from 1999 to 2020.

To evaluate whether the use of two different atmospheric forcings introduced any biases in our results, we conducted additional single-point simulations with GSWP3 and NLDAS forcings specifically for corn and the period from 2001 to 2014. We compared key variables such as soil N_2O, rainfall, temperature, net primary productivity (NPP), and evapotranspiration, as illustrated in Figure 1 below.

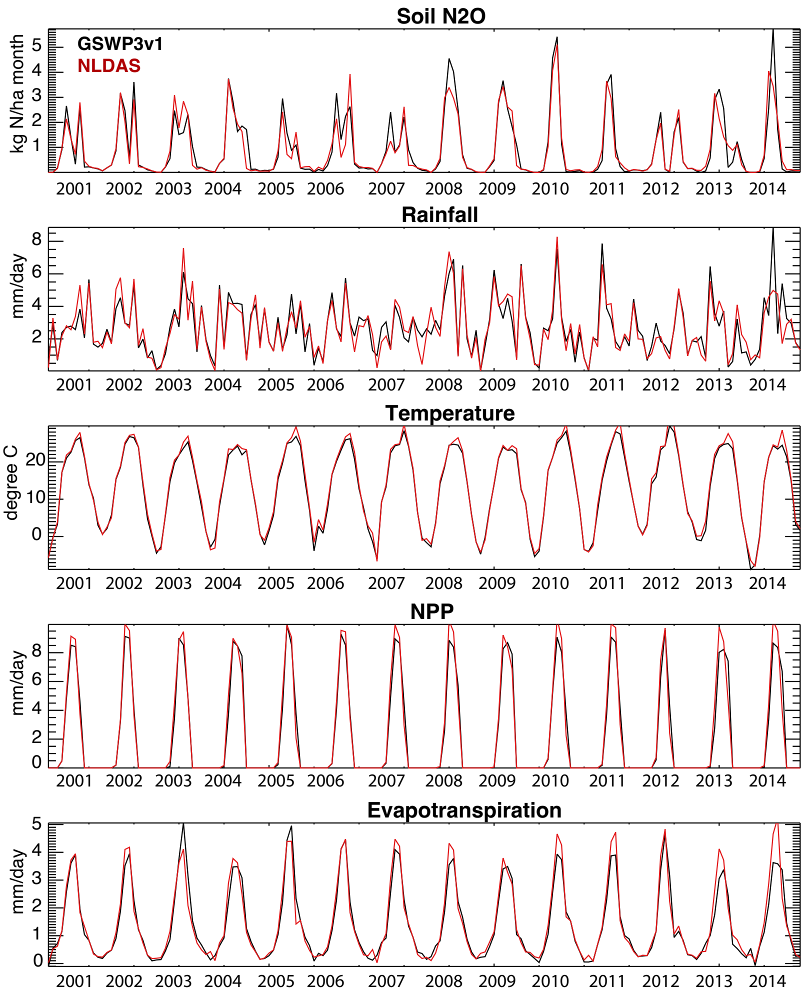

Figure 1. Monthly timeseries of soil N2O, precipitation, temperature, evapotranspiration and NPP for single-point simulation for corn at the Energy Farm, from 2001 to 2014 with NLDAS (red) and GSWP3v1 (black) atmospheric forcings.

Notably, our analysis revealed no significant changes in vegetation and soil dynamics resulting from the use of these different forcings. We recognize the importance of this concern and have added a note in the manuscript to address it accordingly.

Section 2.3 Lines 228-230

> Following the historical simulation, the Energy Farm simulations were run from 2015 to 2019 with meteorological forcing data retrieved from the North American Land Data Assimilation System (NLDAS) forcing dataset (Xia et al., 2012), and initial conditions starting in 2015 for the two single-point simulations, without basalt ('Control' Run) and with basalt ('ERW' Run) application. The use of two different atmospheric forcings, GSWP3 (1901-2014) and NLDAS (1999-2020), was necessary in this study due to their distinct time coverage. Although this approach has the potential to introduce biases and changes in soil dynamics, we conducted a comparison for a coincidental period (2001-2014) and found no significant impacts on vegetation, soil nitrogen fluxes and soil dynamics (Fig. S4 in SM).

*New development introduced a lot of constant parameters (e.g., fN2Onit = 721.86*e^-2.387*pH). fN2Onit has two constant parameters: 721.86, 2.387. These parameters were derived from previous studies, however, also had large uncertainties. And their incorporation into a global model like CLM, will have corresponding parametric uncertainty. How do we confidently know the constant parameters work well at global scale? A common strategy in previous CLM development is to define parameter ranges and conduct model tuning for newly introduced parameters. In such way, we will not only improve the model performance, but also gain knowledge about model parametric uncertainties.*

We acknowledge the reviewer's valid concern regarding the need for a comprehensive sensitivity analysis to assess the model's performance and uncertainty on a global scale. Unfortunately, there is no possibility of expanding the sensitivity analysis of N_2O responses to basalt to other sites because those data do not exist. However, in this study we evaluated our model against field observations 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.

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. 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 such as 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.

We acknowledge that certain aspects of our implementations, such as the fN2Onit parameterization, have undergone limited testing on a global scale, as pointed out by the reviewer. However, at typical soil pH levels in croplands (ranging from 5.8 to 6.2), the variation of fN2Onit is within a range of 3 to 7×10^{-4}, which does not deviate significantly from the originally implemented value in CLM5 of 6×10^{-4} (Li et al., 2000).

To assess the sensitivity of the model to this specific parameterization, we conducted a sensitivity analysis using a single-point setup at the Energy Farm, with a soil pH of 5.9. This setup was designed to represent typical soil conditions found in croplands, allowing us to evaluate the model's performance and response in specific cropland environments, which aligns with the focus of our study. We perturbed the fN2Onit parameter by ±20% and observed that the overall effect on soil N_2O was minimal, ranging from 0.04% to 0.3%.

We clarified this in the manuscript as follows:

Section 2.2.1. Lines 139-143

> The updated fN_2O_{nit} function made the nitrification rate in CLM5 go from the global constant average of 0.06% to 0.3% and increased the global N_2O nitrification/denitrification ratio from 1% to 14%, more accordingly to previous estimates (Inatomi et al., 2020). It should be noted that fN_2O_{nit} values at typical soil pH levels in croplands (5.8 to 6.2) fall within a relatively narrow range of 3 to 7×10^{-4}, which is not significantly different from the original 6×10^{-4} implemented in the model. Small variations in fN_2O_{nit} (e.g., ±20%) have a negligible impact on the total soil N_2O fluxes, with changes ranging 0.04 to 0.3%. However, further work is needed to evaluate the sensitivity of the model to this specific parameterization under other soil conditions, as well as to incorporate the influence of other environmental factors, such as water content and temperature.

Furthermore, we have taken care to emphasize in the manuscript that our implementations represent an initial attempt at parameterizing ERW in a land model, and that ongoing efforts are necessary to refine the ERW schemes and enhance the nitrogen cycling in CLM5 as additional experimental data become accessible (as discussed in our response below).

How CLM5 estimate the dynamics of soil pH is missing? A brief description of the weathering module and how that affect soil pH is needed in the methodology section. Also, a comparison and discussion of observed versus modeled pH variability is needed to demonstrate that CLM5 was able to capture reasonable pH dyanmics after e.g., applying the crushed silicate rock.

We appreciate the reviewer's observation regarding the clarity of our weathering module description. The weathering module in CLM5 enables the activation of weathering, specifically the modification of soil pH through the application of basalt. This functionality is achieved by CLM5 reading the offline changes in soil pH generated by the LC3M ERW model. Essentially, CLM5 obtains spatially distributed delta pH values corresponding to basalt application on crops, thereby adjusting the initial soil pH baseline.

While it is ideal to validate the modelled changes in soil pH resulting from basalt application, data availability for such validation is currently limited, as mentioned above. Field trials investigating ERW are scarce worldwide, and some of the existing data are not yet publicly accessible. In our specific LC3M field trials conducted at the Energy Farm (US Corn Belt 2016-2020), the maize and soybean plots exhibited soil pH values ranging from 6 to 6.5, which align with the soil pH values of 6-7 provided by the Harmonized World Soil Database at a 0.9x1.25 horizontal resolution (Figure S1 in Supplementary Material). Following a 4-year basalt treatment, soil pH increased by approximately 1 unit at the 0-10 cm depth and 0.5 units at the 10-30 cm depth These results are part of an additional study and currently under review.

Notably, the ERW model predicts a change in soil pH of 1.2-1.6 units at the specific location of the Energy Farm after a 25-year treatment (Figure S5 in Supplementary Material).

We understand the importance of validating these soil pH changes resulting from basalt application. However, as mentioned above due to the limited availability of comprehensive field trial data, particularly in relation to long-term studies, obtaining precise validation for the specific pH modifications is challenging. We are actively working on further research and collaboration to enhance the validation of these soil pH changes.

We have clarified the description of the weathering module in the revised manuscript accordingly.

Section 2.3 Lines 242-245

> To model the effect of basalt addition on the N_2O, NO and NH_3 fluxes from soil, we developed a weathering ~~module~~ option for CLM5, in which ~~dynamic~~ 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.

Site level simulation is convincing, but the global comparison needs to be improved. Based on the model performance shown in Figure 6, Table 2, the simulated changes in N2O/NO/NH3 fluxes (figure 7) were not convincing, therefore, the major conclusion of ~30% reduction in N2O/NO emission due to ERW was not strongly supported. Overall, I think this study is timely and important, but the model seems not ready for a global application.

In response to a suggestion by Reviewer 2, we have thoroughly reassessed our global comparison (section 3.2) to enhance the clarity and temper the optimism of our model results. Throughout the review process, we identified a bug in one of our scripts that affected the calculation of soil nitrogen fluxes presented in Tables 2-4, as well as the comparison of soil NH_3 against emission inventories (Figures 6 and S8). However, we want to emphasize that despite this issue, the primary outcomes and conclusions of our manuscript remain unchanged. In fact, the updated values have resulted in an improved alignment with the global emission inventories. We have updated the text to reflect these changes, accordingly.

We respectfully disagree with the reviewer's perspective regarding the changes in soil N_2O, NO, and NH_3 not being convincing. We acknowledge that further efforts are necessary to improve the nitrogen cycling representation in CLM5, as highlighted by Nevison et al. (2022b). Additionally, the implementation of enhanced rock weathering in global models would benefit from further improvements, especially as more field trial data become available. It is important to note that the primary objective of this study is to present an initial implementation of these processes and to analyze the differences between a Control and ERW scenario to tease out the unintended consequences of basalt application in crops. Any model biases are expected to offset each other, and they are unlikely to significantly impact the primary conclusions drawn from the modelling experiment.

We have added the following discussion in Conclusions to acknowledge the need for further work.

Section 5 Lines 512-518

> We acknowledge the need for further improvement in the CLM5 nitrogen cycling representation and the ERW parameterizations. In a comprehensive evaluation of CLM5 nitrification and denitrification processes, Nevison et al. (2022b) emphasized that the

nitrification:denitrification ratio (2:1) in CLM5 is likely to be unrealistically low, even when considering the missing N mineralization term in potential nitrification (Section 2.2.1). Consequently, CLM5 underestimates the fraction of gross mineralization leading to nitrification and overestimates NH_4^+ uptake by plants. Additionally, CLM5 underestimates NO_3 assimilation by immobilizing bacteria. To enhance the confidence in our land model simulations, it is thus crucial to gather more experimental data from ERW field trials as well as observational constraints on soil nitrogen fluxes and flux ratios.

Our study represents a first implementation of an ERW parametrization in a land model N cycling, which has enabled us to understand the implication of large-scale deployment of ERW with croplands on direct soil nitrogen trace gas emissions.