<u>Response to Referee #1</u>

We express our gratitude to Referee #1 for his/her useful comments. Our response to the reviewer's comments and the corresponding revision are described in detail and separately below. The numbers of pages, lines, equations, tables and figures are those in the revised manuscript unless otherwise described.

<u>Comment 1</u>:
"Similar models have been implemented since about 30 years. I think that the authors forgot a few references. For examples, ref to the SAFE and PROFILE box-models should appear in the text, because these are the first to apply laboratory kinetic laws to the field. The WITCH model has been also developped to simulate weathering reactions mechanistically, and a version of it includes the CO2 diffusion (check Goddéris et al., 2006, in GCA; Roland et al., 2013 in Biogeosciences; Beaulieu et al. 2012 in Nature Climate Change; Goddéris et al., 2013 in Biogeosciences). Although not dealing with enhanced weathering, these simpler box-models demonstrate their ability to simulate weathering processes in the field based on a more mechanistic formulation."

Response:
We recognized the significance/importance of developments and successful applications of the already published models including the SAFE, PROFILE, and WITCH models, for a better understanding of ongoing natural weathering and predicting the future weathering in a process-based manner. We did not refer to them because we focused on simulations of enhanced rock weathering, i.e., application of crushed rocks and reactive transport of them at the soil surface, which most of the models were not specifically designed for. Nonetheless we agree with the reviewer that it is important to mention these models as they and our model have a shared goal of a better understanding of weathering processes in general.

Changes in manuscript (Page numbers/Line numbers):
We added a sentence to refer to the previous models (P2/L48-53).

<u>Comment 2</u>:
"I found a bit strange to talk about uplift. Is the model really accounting for uplift ? If I understand it correctly, it seems to me that the model is using a referential in which fresh primary minerals are going upwards as weathering proceeds. In the real world, it is the regolith which progressively penetrates the bedrock and thus goes downward. This has nothing to do with uplift ?"

Response:

We utilized a spatially fixed frame for the model's calculation domain and solid phases can be supplied from the bottom via uplift from a deeper depth and can be lost via erosion at the surface after the reaction and transport within the model domain. This is a common assumption taken in many 1D reactive-transport models and consistent with the dynamics of rocks in the real world (e.g., Bolton et al., 2006; Li et al., 2014). We believe that the reviewer referred to the propagation of reaction front, which can be related to the interface between soil/regolith and parent/bed rocks, as they deepen with time as inferred from the reviewer's comment. Our model can indeed simulate deepening of reaction front with time (e.g., Figs. 1-3) within our calculation domain. Please also note that steady state for solid phases (and thus reaction fronts and/or bedrock/regolith interfaces) can be achieved within a spatially fixed frame only when considering the solid phase supply from the depth (uplift); otherwise, there is no source to balance the loss via dissolution/erosion, assuming no external supply (e.g., dust). The effect of including uplift and erosion may not be significant if we consider reaction/transport only on anthropogenic timescales (e.g., < 100 years), but we enabled solid phase erosion as well as uplift to make the model comprehensive and applicable for tuning at different and flexible timescales, which can be longer than the anthropogenic timescale (e.g., Fig. 13). Enabling erosion can also be justifiable or even desirable, given the enhancement of soil erosion at croplands (e.g., Pimentel, 2006, Environ. Dev. Sustain, 8, 119).

Changes in manuscript (Page numbers/Line numbers):
Because we have already described uplift/erosion with references in lines 88-90, we did not make any changes regarding explanations of uplift/erosion. However, we added a sentence in order to more clearly define our calculation domain (P3/L92-P4/L94).

Response to Referee #2

We express our gratitude to Referee #2 for his/her useful comments. Our response to the reviewer's comments and the corresponding revision are described in detail and separately below. The numbers of pages, lines, equations, tables and figures are those in the revised manuscript unless otherwise described.

Comment 1:

"l. 15. Suspect the authors of the Beerling et al. would quibble with the notion it lacked mechanistic detail. Theirs was a 1-D model calibrated against a phreeqC RTM of comparable to detail to the present one. That strategy gave great flexibility both for integrating into a broader EW techno-economic framework and future development in terms of adding geochemistry details and year-to-year particle size treatment."

Response:

We agree with the reviewer that the Beerling et al.'s approach is detailed and novel. However, we consider that their modeling approach still lacks some mechanistic aspects of enhanced rock weathering simulations as detailed below.

Beerling et al. (2020) used PhreeqC 1D simulation to calibrate their 'performance' model for the global application. Either in their benchmark PhreeqC 1D or their 'performance' simulations, soil C is not explicitly simulated, nor does the model simulate soil mixing of solid phase components of crushed and applied basic rocks. Also, PhreeqC 1D simulations couple reaction and transport in a time-explicit way if one uses the time-explicit 1D transport algorithm in PhreeqC. This requires fine time steps to avoid numerical errors, which may not be a problem for short-timescale simulations (e.g., <100 years) but can become problematic if one wants to spin up the model, e.g., to reproduce background conditions before a basalt application experiment on a timescale longer than e.g., anthropogenic timescale. In addition, although their 'performance' model implements PSD tracking, they did not provide detailed results regarding their calculated PSDs to which other models (including ours) can be compared. We attempt to directly address these mechanistic details. For example, our model enables application and soil mixing of variable solid phases (including soil OM); our model fully couples reaction and transport where concentrations of all species in a given time step are time-implicitly solved as unknowns satisfying the mass balance reflecting the reactions and transport at once (e.g., Steefel and Lasaga, 1994); and we illustrated how the PSDs are tracked with our model in detail from theory to numerical scheme to results.

One advantage of using PhreeqC is that thermodynamic database is more easily and self-consistently switched/chosen. This aspect will be addressed in the future release of our model.

Changes in manuscript (Page numbers/Line numbers):
We modified the sentence to be clearer (P1/L14-16).

Comment 2:
"l. 47-48. Agreed. Ensembles of traceable EW models for CDR estimate will be an important advance in this field."

Response:
We agree on the reviewer's point.

Changes in manuscript (Page numbers/Line numbers):
We made no changes in manuscript in response to the comment.

Comment 3:
"l. 480. Treatment of biotic weathering seems to be mainly through the production of SOM and its subsequent decomposition, as it affects the acid-base balance. But there is also the effect of nutrient uptake (base cations) by plant and release of protons by fine roots likely to be in direct contact with silicate rock grains, and production of organic acids by fine and mycorrhizal fungi which also produce a focused release of H+ from explorative hyphae (this is partially noted in the conclusions but the omission might be flagged here too)."

Response:
We agree with the reviewer that biotic effect on weathering is not limited to SOM production and decomposition. Indeed, we are planning to implement uptake of cations/nutrients by plants by coupling SCEPTER with a previously developed soil ecosystem model. Another less explicit way to implement cation/nutrient uptake by plants would be to enable cation exchange in SOM, which has been done in the developing version of our code. The developing version of the model has also enabled tracking of oxalic acid including its decomposition, proton releases, and complexation with cations (e.g., Lawrence et al., 2014, GCA 139, 487; Perez-Fodich and Derry, 2019, GCA 249, 173). CO_2 production via SOM respiration is still likely to be a dominant biotic factor in natural weathering (e.g., Perez-Fodich and Derry, 2019; Beerling et al., 2020) and thus one can regard the current version as the model that already has implemented the basics for biotic weathering. We are planning to release an updated version that includes plant uptake of nutrients and secretion of organic acids in the near future.

Changes in manuscript:
We described additional biotic effects in the relevant section (P18/L492-494) as well as in the

conclusion section (P22/L645-46).

Comment 4:

"l. 615. Part of the explanation in the timescale of CDR efficiency between the present model and Beerling et al. may be related to the treatment of particle size distributions in the soil from one year to the next (discrete vs continuous)."

Response:

We agree with the reviewer that the difference in the capture outcome is likely at least partly attributable to the difference in the PSD calculation between the present model and the model of Beerling et al. We now suggest in the text that a model intercomparison study would be very illuminating (please also see our response to Comment 1 by the reviewer).

As for the time step, so long as it is small enough the difference between numerical and analytical solutions should be minor. As the model adopts relatively fine timesteps for the ERW experiments (mostly <0.01 yr), we do not consider that taking discrete time steps in our model would be the main cause of the difference between our and the Beerling et al.'s models. As we partly discussed in response to Comment 1 by the reviewer, there are many other factors that this difference could be attributed to. For instance, our PSD calculation accounts for addition and mixing of added particles within the mixed layer, while it seems that the PSD changes are caused only as a result of dissolution in Beerling et al. (2020). Moreover, experimental setups are quite different between the two models. We first spin up the model to reproduce the porewater pH and SOM distributions (e.g., Fig. 13) and then start basalt application experiments as restarts from spin-up, while the spin-up phase does not exist in the experimental setup by Beerling et al. (2020). As we stated in the relevant section, C capture during ERW experiments depends significantly on the soil environments and thus on the spin-up phase before the basalt application (e.g., Figs. 14 and 15). Overall, we consider that we need an intercomparison study to conclude upon the difference between the two models where experimental setups, boundary conditions and model parameterizations are aligned between the models as much as possible.

Changes in manuscript:

We added a statement that we need an intercomparison study to conclude upon the difference between the models (P22/L630-633).