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Advancing Enhanced Weathering Modeling in Soils: Critical Comparison with Experimental Data

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Key Points:

- We present a depth-averaged, dynamic Soil Model for Enhanced Weathering (SMEW).
- The model results are critically compared with four experimental datasets of different complexity.
- The comparison demonstrates slower-than-expected dissolution rates.

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Abstract

Enhanced weathering (EW) is a promising strategy to remove atmospheric CO₂ by amending agricultural and forestry soils with ground silicate rocks. However, current model-based EW assessments face large uncertainties stemming from the intricate interplay among soil processes, compounded by the absence of a detailed comparison with available observational data. Here, we address this critical gap by first advancing a dynamic, eco-hydrological, and biogeochemical Soil Model for Enhanced Weathering (SMEW). We then conduct a hierarchical model-experiment comparison with four experimental datasets of increasing complexity, from simple closed incubation systems to open mesocosm experiments. The comparison demonstrates SMEW's ability to capture the dynamics of primary variables, including soil moisture, alkalinity, and inorganic carbon. The comparison also reveals that weathering rates are consistently lower than traditionally assumed by up to two orders of magnitude. We finally discuss the implications for carbon removal scenarios and avenues for further theoretical and experimental explorations.

Plain Language Summary

Enhanced weathering (EW) is a promising strategy to mitigate climate change while increasing agricultural productivity and mitigating ocean acidification. The strategy involves amending cropland and forest soils with finely ground silicate rocks, which sequester atmospheric CO₂ upon dissolution. However, current EW assessments relying on models face uncertainty, primarily stemming from challenges in accurately representing the intricate hydrological and biogeochemical processes driving mineral dissolution in the soil. The absence of a robust model-data comparison exacerbates these uncertainties. This study addresses these issues by presenting a model for EW dynamics in the upper soil layer, successfully replicating diverse experimental datasets. Our model reveals a slower mineral dissolution than conventionally assumed, offering insights into EW potential as a negative emission strategy.

1 Introduction

In addition to emissions reduction from every sector, significant carbon dioxide removal (CDR) through negative emission technologies (NETs) is needed to limit global warming (Calvin et al., 2023). Among various proposed NETs, enhanced weathering (EW) is emerging as one with considerable CO₂ removal potential and low technological requirements (Kohler et al., 2010; Renforth, 2012; Berge et al., 2012; Hartmann et al., 2013; Taylor et al., 2016; Beerling et al., 2020; Calabrese et al., 2022). EW relies on amending agricultural and forestry soil with crushed silicate materials (e.g., basalt, dunite, wollastonite) to promote biomass growth and sequester CO₂ in aqueous or mineral forms (Hartmann et al., 2013; Taylor et al., 2021). Upon dissolution, the hydrologic cycle transports part of the EW products to surface freshwaters and the ocean, mitigating ocean acidification and stably sequestering atmospheric CO₂ for geological timescales (Renforth & Henderson, 2017; Bertagni & Porporato, 2022). As a further co-benefit, EW is expected to improve nitrogen use efficiency (NUE) in agricultural fields, reducing reactive nitrogen emissions and the demand for fossil-fuel-based fertilizers (Blanc-Betes et al., 2021; Val Martin et al., 2023). Deployed over suitable lands at the global scale, it is estimated that EW potential may reach the order of gigatonnes of CO₂ removal per year (Taylor et al., 2016; Strefler et al., 2018; Beerling et al., 2020; Baek et al., 2023).

Despite these promising estimates, EW assessments face significant uncertainties rooted in the complex interplay between hydrological and biogeochemical processes across scales (Calabrese et al., 2022). Weathering rates exhibit considerable variability, spanning orders of magnitudes due to rock specifics, and temporal and spatial heterogeneity in hydroclimatic drivers and soil processes (Jung & Navarre-Sitchler, 2018; Li et al., 2022; Schabernack & Fischer, 2022; Deng et al., 2022). This variability complicates ef-

67 forts to reconcile theoretical expectations with laboratory observations, even for the same
68 rock type (Renforth et al., 2015; Amann et al., 2020; Buckingham et al., 2022). Field
69 trials are in their nascent stages, and monitoring, reporting, and verifying (MRV) car-
70 bon dioxide removal in open, heterogeneous, and multiphase soil systems face inherent
71 challenges (Clarkson et al., 2023), although recent efforts have started to address them
72 (Amann & Hartmann, 2022; Reershemius et al., 2023; Knapp et al., 2023). Specifically,
73 soil-based mass balance approaches, initially used for natural chemical weathering but
74 modified to reduce analytical error, could be a promising option to resolve the small signal-
75 to-noise ratios in EW mineral cation depletion (Reershemius et al., 2023). When EW
76 is applied to significant portions of a watershed area, stream water chemistry analyses
77 may also be beneficial to understand the transport of the mineral dissolution products
78 (Larkin et al., 2022), as in the case of agricultural liming (Hamilton et al., 2007). Quan-
79 tifying this transport from the field to CO₂ storage locations, such as deep aquifers or
80 the oceans, is a crucial and yet largely unexplored EW aspect (Hartmann et al., 2013;
81 Zhang et al., 2022; Bertagni & Porporato, 2022; Calabrese et al., 2022; Bertagni et al.,
82 2024).

83 Within this intricate context, current estimates of EW potential as a NET heav-
84 ily rely on models, mostly vertically explicit reactive transport models, where minerals
85 added to the topsoil layers undergo dissolution based on transition state theory (Taylor
86 et al., 2016; Beerling et al., 2020; Kantzas et al., 2022; Baek et al., 2023). While these
87 models can comprehensively treat reacting chemical species across a heterogeneous soil
88 profile, they are usually used under simplifying assumptions, such as constant vertical
89 water flow and homogeneous soil properties. An alternative, spatially lumped approach
90 focuses on the temporal dynamics of average quantities within the upper soil layers – the
91 most dynamic soil layers, where ground rocks are introduced – hence emphasizing tem-
92 poral over spatial variability (Cipolla et al., 2021a, 2021b). Given the direct and indi-
93 rect impacts of hydroclimate conditions and soil moisture dynamics on weathering rates,
94 capturing temporal variability is crucial. Soil moisture influences the surface area of EW
95 material in contact with water and the dilution and leaching of weathering products. More-
96 over, it affects biotic and abiotic soil processes influencing EW dynamics, including car-
97 bon cycling, bacterial activity, and solute, heat, and gas transfers (Manzoni et al., 2012;
98 Miele et al., 2023; Porporato & Yin, 2022).

99 Surprisingly, despite numerous experimental works in the last decade (Dietzen et
100 al., 2018; te Pas et al., 2023; Kelland et al., 2020; Amann et al., 2020; Renforth et al.,
101 2015; Vienne et al., 2022; Buckingham et al., 2022), comparisons of model results with
102 experimental observations have been minimal (Kelland et al., 2020). An extensive model-
103 data comparison is hence pivotal, not only to validate EW models for realistic assess-
104 ments of net-zero scenarios but also to improve modeling assumptions, provide a hypothesis-
105 testing tool to investigate EW processes, design better experiments, and quantify un-
106 certainty.

107 Our work addresses this gap, presenting an ecohydrological and biogeochemical Soil
108 Model for EW (SMEW) and using the model for an extensive and systematic model-experiment
109 comparison. Specifically, the model is a substantial evolution of the model initially con-
110 ceived by Cipolla et al. (2021a), including several improvements in model closures and
111 adding new model components (Sec. 2). The model performance is then compared with
112 four experimental datasets covering a gradient of complexity, from simple closed incu-
113 bation systems to more complicated, open mesocosm experiments (Sec. 3). The model-
114 data comparison demonstrates that the model captures the dynamics of the primary vari-
115 ables of interest and provides crucial insights into weathering rates (Sec. 4). We finally
116 identify model limitations and discuss areas requiring further theoretical and experimen-
117 tal exploration.

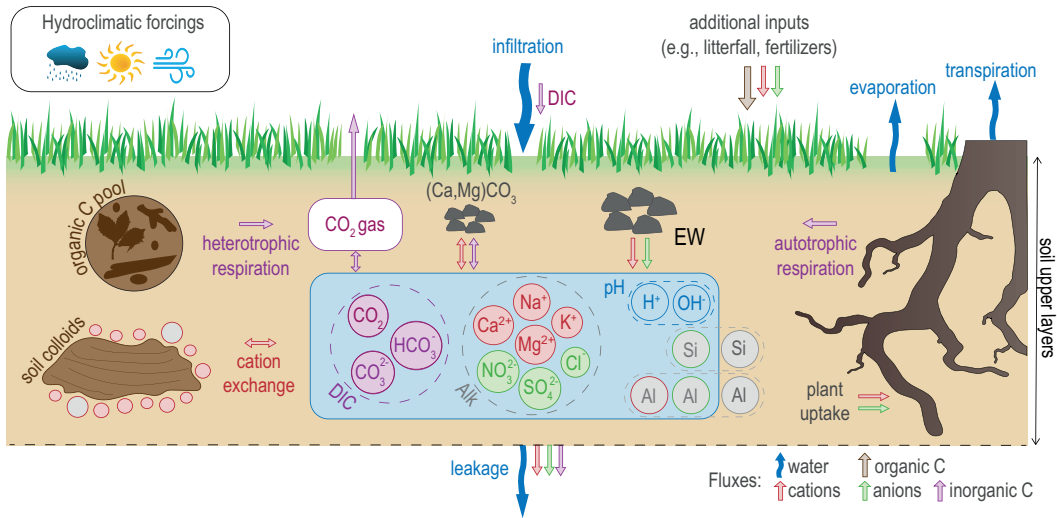


Figure 1. Sketch of the biogeochemical and ecohydrological processes represented in the Soil Model for Enhanced Weathering (SMEW). SMEW is a dynamic, depth-averaged model for the upper soil layers where the crushed rock is applied.

2 Soil Model for Enhanced Weathering (SMEW)

This model builds upon prior work on EW in the soil's upper layers (Cipolla et al., 2021a, 2021b) incorporating several extensions and improvements in model closures and new model components. The model emphasizes the dynamic behavior of depth-averaged quantities within the root zone. The depth-averaged approach is especially appropriate when agricultural practices have homogenized the upper soil layers (Porporato & Yin, 2022). The critical components of the model revolve around the intricate interplay between the water balance, influenced by stochastic infiltration rates, and the biogeochemical processes occurring within the multiphase soil porous media. Fig. 1 provides an overview of the main model components. In this section, we discuss the mass balances for the key variables of interest, which form a dynamic system of ordinary differential equations (ODEs). Jointly with the ODE system, we solve a set of algebraic equations based on a quasi-steady-state approximation to account for aqueous carbon and aluminum speciation and the cation redistribution between adsorbed and dissolved phases (Appendix A). The dynamics of plants and their roles in EW are presented in Appendix B. A discussion of the model parameters is reported in the Supporting Information (Text S1).

2.1 Hydroclimate and Moisture Dynamics

Hydroclimatic forcings such as temperature and rainfall exert critical controls on weathering rates (Calabrese & Porporato, 2020; Deng et al., 2022) by directly influencing water availability and distribution, mineral dissolution kinetics and impacting various biogeochemical processes, including biotic activity and chemical equilibria. In SMEW, these hydroclimatic factors can be introduced through observational or reanalysis data or generated through modeling for future projections. Of particular significance to the correct representations of weathering dynamics are the short-term hydrological fluctuations because of their nonlinear feedback on soil hydro-biogeochemistry (Laio et al., 2001; Porporato, D'Odorico, et al., 2003; Cipolla et al., 2021a; Porporato & Yin, 2022; Dong et al., 2023). Consequently, our modeling framework incorporates a water mass balance

reproducing the time (t) evolution of relative soil moisture (s) within the soil depth (Z)

$$nZ \frac{ds}{dt} = R(t) - Q(s, t) - E(s) - T(s) - L(s), \quad (1)$$

135 where n is soil porosity, $R(t)$ is rainfall, $Q(s, t)$ is runoff, $E(s)$ is evaporation, $T(s)$ is plant
 136 transpiration, and $L(s)$ is leaching. In the absence of data, rainfall can be modeled as
 137 a stochastic marked Poisson process (Rodríguez-Iturbe et al., 1999; Porporato & Yin,
 138 2022). Surface runoff, resembling Horton overland flow, is activated when the rainfall
 139 exceeds the available storage capacity. Evaporation and transpiration fluxes are influ-
 140 enced by soil moisture and vegetation cover (Laio et al., 2001), and their cumulative ef-
 141 fect is bounded by the potential evapotranspiration (ET_0), which is estimated using the
 142 Penman-Monteith method for a reference crop, based on climatic conditions such as tem-
 143 perature, wind speed, latitude, and albedo (Allen et al., 1998). Water leakages to lower
 144 soil horizons are modeled as a power law of soil moisture with coefficients depending on
 145 soil texture (Laio et al., 2001).

146 2.2 Organic Carbon and Heterotrophic Respiration

In the topsoil layers, soil carbon exists in organic and inorganic forms. The flux from organic to inorganic carbon pools results from the decomposition of soil organic matter, mostly driven by biotic processes like bacterial activity. This flux, called heterotrophic soil respiration, is a key contributor to elevated CO_2 levels in soil air, making it a critical factor in the potential EW efficiency in soil carbon sequestration. To model the dynamics of organic carbon (OC), here considered as dead biotic material, we employ a simple balance that includes an addition term (ADD), accounting for inputs like litterfall or soil amendments, and a decomposition term (DEC) representing biotic activity (Porporato, D’Odorico, et al., 2003; Cipolla et al., 2021a). The mass balance for OC is

$$\frac{d\text{OC}}{dt} = \text{ADD} - \text{DEC}. \quad (2)$$

147 Depending on the available information, the addition of OC can be assumed to be con-
 148 stant, vary seasonally, or be based on photosynthetic activity. The decomposition term
 149 is proportional to the available OC through a moisture- and temperature-dependent co-
 150 efficient (Porporato, D’Odorico, et al., 2003; Cipolla et al., 2021a). A fraction r of the
 151 decomposed OC is converted into inorganic carbon, defining soil heterotrophic respira-
 152 tion ($\text{RESP}_h = r \text{DEC}$). The remaining fraction ($1 - r$) is assumed to be converted
 153 into living biomass of soil biota (e.g., bacteria, fungi, and soil fauna), which is not ex-
 154 plicitly modeled (Porporato, Laio, et al., 2003). While here we use a minimalist soil OC
 155 model, more elaborate representations of the OC cycle (e.g., with explicit microbial dy-
 156 namics) may be adopted (Wieder et al., 2013; Jha et al., 2023) based on specific scien-
 157 tific questions being addressed in the interactions between OC and EW.

158 2.3 Inorganic Carbon Pools

EW negative-emission potential hinges on the sequestration of inorganic carbon, mainly in the form of aqueous carbonates within soil water and throughout the hydrological cycle, or through the formation of secondary carbonate minerals, albeit with a 50% reduction in CO_2 removal efficiency (Hartmann et al., 2013; Bertagni & Porporato, 2022). The main components of soil inorganic carbon include CO_2 in the soil air phase, dissolved inorganic carbon (DIC) in the soil water, and carbon stored in mineral forms. Given that the equilibration timescale of aqueous and gaseous forms is much faster than that of carbonate mineral precipitation and dissolution, we consider two distinct inorganic carbon pools: one that combines aqueous and gaseous forms (IC) and another accounting for mineral inorganic carbon (MIC). The overall mass balances for IC and MIC

are expressed as:

$$\frac{dIC}{dt} = \text{RESP}_{\text{h+a}} + I_w \cdot [\text{DIC}]_{I_w} - L \cdot [\text{DIC}] - F_{\text{ADV+DIFF}} + W_{(\text{Ca, Mg})\text{CO}_3}, \quad (3)$$

$$\frac{d\text{MIC}}{dt} = -W_{(\text{Ca, Mg})\text{CO}_3}, \quad (4)$$

159 where $[\cdot]$ indicates concentration. $\text{RESP}_{\text{h+a}}$ is the sum of heterotrophic and autotrophic
 160 respiration, respectively. Autotrophic respiration (RESP_{a}), namely the release of CO_2
 161 gas by plant roots, is estimated to scale with vegetation (Appendix B) and to be equiv-
 162 alent to heterotrophic respiration (Sec. 2.2) when plants are fully grown (Bond-Lamberty
 163 et al., 2004). Another minor source of inorganic carbon in the soil is the DIC in infil-
 164 trating water ($I_w = R - Q$). IC can exit the control volume as aqueous DIC through
 165 leaching ($L \cdot [\text{DIC}]$) or as gaseous CO_2 to the atmosphere through diffusive or advec-
 166 tive fluxes ($F_{\text{ADV+DIFF}}$) (Millington & Quirk, 1961; Cipolla et al., 2021a). The term $W_{(\text{Ca, Mg})\text{CO}_3}$
 167 accounts for the dissolution (> 0) or precipitation (< 0) of calcium and magnesium car-
 168 bonates, modeled following Kirk et al. (2015). The redistribution of IC between soil air
 169 CO_2 and aqueous carbonates follows equilibrium assumptions (Appendix A).

170 2.4 Biogeochemistry of Alkaline and Acid Elements

To promote inorganic carbon sequestration, EW aims to release alkaline cations
 (Ca^{2+} , Mg^{2+} , K^+ , Na^+) in soil water and throughout the hydrological cycle (Hartmann
 et al., 2013). These cations increase water alkalinity and promote a transfer of CO_2 from
 the atmosphere to the water by forming aqueous carbonates in favorable water-chemistry
 conditions (Bertagni & Porporato, 2022). We hence consider four mass balances for each
 of these alkaline cations, indicated generically as X. The mass balances for any total cation
 content (X_{tot}) within the control volume, comprising cations dissolved in the soil solu-
 tion and those adsorbed onto soil colloids, can be written as

$$\frac{dX_{\text{tot}}}{dt} = I_X + \text{EW}_X + W_{X\text{CO}_3} - (L + T)[X] - \text{UP}_X. \quad (5)$$

171 I_X accounts for background cation inputs like litterfall decomposition, fertilizer addition,
 172 and background weathering processes. EW_X denotes the cation release by the EW ap-
 173 plication, and $W_{X\text{CO}_3}$ is the release from the weathering of Ca or Mg carbonate. The
 174 term $(L+T)[X]$ characterizes the outflow resulting from leaching and plant passive up-
 175 take, while UP_X pertains to active plant uptake during growth (Appendix B). Given the
 176 total cation amount in the control volume, the partitioning between adsorbed and aque-
 177 ous components follows equilibrium assumptions (Appendix A).

A similar mass balance approach applies to the major strong anions commonly found
 in soil solutions (e.g., Cl^- , NO_3^{2-} , SO_4^{2-}) that do not undergo speciation at pH val-
 ues of interest. Conveniently, we do not need to discriminate between the various anions
 of the strong acids because i) EW aims to increase cation concentrations, ii) anion ad-
 sorption is mostly negligible in many soil environments, iii) it is the cumulative presence
 of these anions that defines soil water alkalinity (Appendix A). We can thus collectively
 denote these anions as An and write a single mass balance

$$\frac{d\text{An}_{\text{tot}}}{dt} = I_{\text{An}} - (L + T)[\text{An}], \quad (6)$$

178 where I_{An} signifies background anion input, and $(L+T)[\text{An}]$ represents anion losses due
 179 to leakages and passive plant uptake.

180 2.5 Silicon and Aluminum Balances

Since the most promising options for large-scale EW applications are silicate min-
 erals and rocks, EW is anticipated to release large amounts of silicon (Si) into soils. This

constitutes a potential EW co-benefit, as soil Si is a biotic nutrient that enhances plant immune system (Fauteux et al., 2005; Kim et al., 2014), although the overall effect on soil properties (e.g., hydraulic conductivity) is largely unknown. Soil Si chemistry is rather complex, comprising dissolved species, amorphous solid phases, and organic and inorganic complexes (Schaller et al., 2021), and its dynamics is expected to impact soil formation processes on long timescales (Weil & Brady, 2016). Given that Si impact on the CO₂ sequestration dynamics by EW is minor, here we follow a simplified approach, wherein we consider Si as dissolved silicic acid. More complex mass balances could be adopted depending on the goal of the investigation. The mass balance hence is

$$\frac{d\text{Si}_{\text{tot}}}{dt} = I_{\text{Si}} + \text{EW}_{\text{Si}} - (L + T)[\text{Si}] - \text{UP}_{\text{Si}}, \quad (7)$$

181 where I_{Si} represents background Si inputs, EW_{Si} accounts for Si released through EW
 182 applications, $(L + T)[\text{Si}]$ signifies Si outflow due to leaching and passive plant uptake,
 183 and UP_{Si} denotes active plant uptake during growth.

Aluminum is a prevalent element in highly weathered, acidic soils, where it can be found in complexes, in the soil solution, or as cations adsorbed into soil colloids. In acidic conditions, aluminum plays a crucial role as a buffering agent but can be toxic to soil biotic activity and plants in high concentrations (Weil & Brady, 2016). Although Al is an undesired product, EW applications may release some of it, depending on the mineral composition of the rock applied. The mass balance for aluminum is expressed as

$$\frac{d\text{Al}_{\text{tot}}}{dt} = I_{\text{Al}} + \text{EW}_{\text{Al}} - L[\text{Al}_{\text{mob}}], \quad (8)$$

184 where I_{Al} and EW_{Al} correspond to background and EW-induced Al releases, respectively.
 185 Aluminum losses are assumed to occur solely through the leaching of Al's more soluble
 186 and mobile forms ($[\text{Al}_{\text{mob}}]$), which can be abundant in highly acidic ($\text{pH} < 4.5$) or al-
 187 kaline ($\text{pH} > 7$) conditions. Aluminum speciation reactions are reported in Appendix
 188 A.

189 2.6 Rock Weathering

Modeling rock weathering is pivotal to understanding and quantifying EW dynam-
 ics and temporal efficiency. Applied rocks are typically composed of various minerals,
 and the release of a specific element like an alkaline cation X (Sec. 2.4) results from the
 collective contribution of mineral dissolution. This contribution depends on the mineral
 dissolution rate (W_i) and the mineral surface area (SA_i), and can be expressed as

$$\text{EW}_X = \sum_i m_{X,i} \cdot \text{SA}_i \cdot W_i(s, \Theta, \text{pH}) \quad (9)$$

190 where $m_{X,i}$ accounts for the stoichiometry of the element X in the mineral i , and Θ stands
 191 for temperature. The same equations, with coefficients $m_{\text{Al},i}$ and $m_{\text{Si},i}$, apply to the re-
 192 lease of Al and Si, namely EW_{Al} and EW_{Si} in eqs.(7) and (8). The mineral surface area
 193 SA_i is determined according to the methodology proposed by Beerling et al. (2020), which
 194 accounts for the dynamically evolving rock composition and particle distribution and the
 195 fractal dimension of the particle surface.

For the weathering rates, we follow previous EW modeling efforts (Taylor et al.,
 2016; Beerling et al., 2020; Kanzaki et al., 2022) and use the semi-empirical formula by
 Palandri (2004). This seminal formulation, stemming from the work of Lasaga (1984),
 is based on dissolution experiments in stirred reactors without diffusive limitations and
 under conditions far from equilibrium. The formula accounts for the most well-studied
 mechanisms of mineral dissolution, driven by the water species H₂O, H⁺, and OH⁻. As
 previously implemented by Cipolla et al. (2021a), we also consider the dependence of min-
 eral dissolution rates on the relative soil moisture value (s) to account for the wet por-
 tion of the mineral surface that can actually undergo dissolution. The formula for the

weathering rate of each mineral can thus be expressed as:

$$W_i = F_D \cdot s \cdot \sum_j k_{i,j}(\Theta) \cdot a_j^{n_{i,j}} \cdot (1 - \Omega_i^{p_{i,j}})^{q_{i,j}}, \quad (10)$$

196 where j is the individual weathering agent (H_2O , H^+ , OH^-) and a_j are the agent ac-
 197 tivities (here approximated as concentrations). $k_{i,j}(\Theta)$ are the mineral- and agent-dependent
 198 rates accounting for temperature (Θ) effects, and $n_{i,j}$ are the reaction order constants
 199 (Palandri, 2004). Ω_i is the mineral saturation index (Morel & Hering, 1993). The co-
 200 efficients $p_{i,j}$ and $q_{i,j}$ have been quantified only for very few minerals and agents and are
 201 approximated to unity (Palandri, 2004). F_D is a dissolution factor that we will quan-
 202 tify based on experimental observations. As later discussed (Sec. 4.1), the observation-
 203 driven quantification of F_D is crucial to assess actual weathering rates and the validity
 204 of Palandri’s formulation in the soil environment.

205 2.7 Model Setup and Simulation Example

206 The model requires a characterization of the hydroclimatic forcings, the EW ma-
 207 terial and application rate, the plant, and the soil biophysical properties. Rainfall and
 208 temperature are key hydroclimatic forcings that impact virtually all model components.
 209 Wind speed and albedo play a role in influencing potential evapotranspiration (Allen et
 210 al., 1998). For EW application, details include the amount and mineral composition of
 211 the applied rock, particle size distribution, and specific surface area. Plant character-
 212 ization involves parameters like carrying capacity, growth rate, and root area index, mea-
 213 sured or derived from the literature (Appendix B). Soil characterization requires details
 214 on soil texture, initial organic carbon content and pH, and inorganic chemistry. Notably,
 215 assuming a quasi-steady state equilibrium of the soil chemistry allows constraining the
 216 air-water carbonate system using a single quantity (e.g., the CO_2 partial pressure) in ad-
 217 dition to the soil pH (Stumm & Morgan, 1996). For the biogeochemistry of the major
 218 ions, minimum initial data requires either the adsorbed fractions on the cation exchange
 219 capacity (CEC), the concentrations in the soil solution, or the total quantities per unit
 220 of soil. The others are determined through the Gaines-Thomas convention (Appendix
 221 A). In the absence of specific data, background elemental input fluxes, e.g., I_X in eq. (5),
 222 can be defined to balance background losses, e.g., $[x]_{\text{initial}}(\bar{T} + \bar{L})$ here the bar denotes
 223 temporal averaging. This approach ensures that the initial condition represents a long-
 224 term average state of the soil that is reestablished whenever the EW application is ab-
 225 sent or concluded.

226 Fig. 2 shows an example of a 1-year simulation for a 1 kg/m^2 (10 tonn/ha) EW ap-
 227 plication with forsterite (Mg_2SiO_4) in a temperate humid climate. Longer simulations
 228 of 10 and 50 years showing the long-term impact of mineral dissolution are reported in
 229 Fig. S1 and S2. A baseline simulation without rock application is reported in Fig. S3.
 230 The mean temperature is 13°C , and the yearly cumulative rainfall is 1200 mm. The sim-
 231 ulated soil is an organic-rich (initial OC is 5%) and acidic (initial pH is 4) loam. Results
 232 show how the low soil pH favors the mineral dissolution rate but impedes the formation
 233 of aqueous bicarbonates in the soil solution for the first 170 days. The trend reverses when
 234 the pH gets around 6, with slower dissolution rates and bicarbonate formation. This trade-
 235 off between mineral dissolution kinetics and CO_2 sequestration efficiency has been pre-
 236 viously explored (Bertagni & Porporato, 2022). Additionally, as the pH rises, the CEC
 237 base saturation increases, with Mg^{2+} replacing the acid ion H^+ (panel e), highlighting
 238 the potential of EW for liming purposes.

239 3 Comparison with Experiments

240 This section compares the model outcomes with available experimental data. Specif-
 241 ically, we use four different experimental datasets derived from experiments conducted
 242 at varying levels of complexity: i) small-scale vials open to the atmosphere and with moist

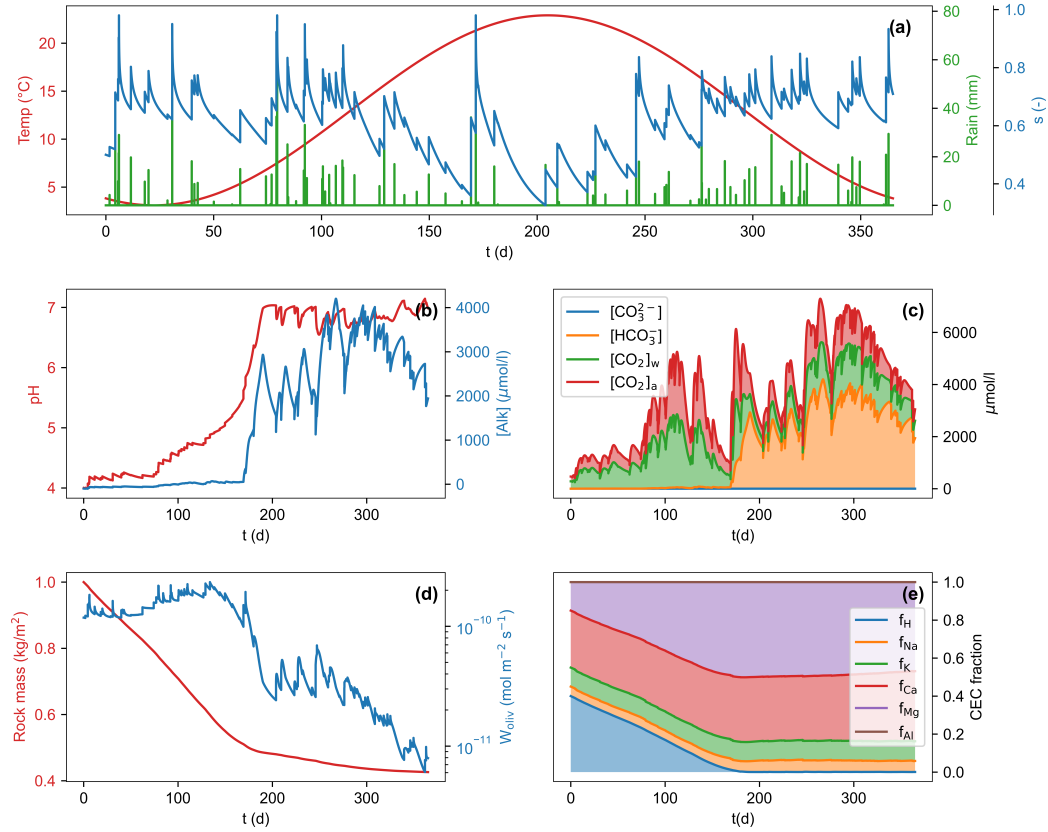


Figure 2. Example of model output for an EW application (1 kg/m^2) with forsterite (Mg_2SiO_4) in a temperate humid climate ($F_D = 1$). (a) Temperature, rainfall, and soil moisture. (b) Soil water pH and alkalinity. (c) Inorganic carbon speciation. (d) Mineral mass and weathering rate. (e) Soil cation adsorption. Simulation results extended to 10 and 50 years are reported in Fig. S1 and S2. A baseline simulation without the rock application is reported in Fig. S3.

243 acidic soils (Dietzen et al., 2018); ii) small down-flow soil columns open to the atmosphere
 244 and water leaching (te Pas et al., 2023); iii) two more complete mesocosm experiments
 245 incorporating growing vegetation (Kelland et al., 2020; Amann et al., 2020). The com-
 246 parison with these different experimental setups gives the advantages of compartmen-
 247 tal investigations of the model performance and a broad examination of EW dynamics
 248 under different environmental forcings. Because our model is spatially lumped and de-
 249 signed for the upper soil layers, we preferred not to include experiments conducted with
 250 vertically deep and heterogeneous soil cores (Renforth et al., 2015; Vienne et al., 2022;
 251 Buckingham et al., 2022). A summary of the model set-up based on available experimen-
 252 tal information is provided in Table S1 in the Supporting Information.

253 3.1 Acidic Soil in Vials

254 Our first comparison involves the experiments by Dietzen et al. (2018), which en-
 255 tailed a three-month soil incubation study to assess the weathering of olivine (mainly
 256 composed of forsterite, Mg_2SiO_4) and its impact on available Mg levels, pH, and soil CO_2
 257 flux. The experiments employed 110 ml open vials filled with soil that remained consis-
 258 tently moist throughout the study. Olivine was added at varying application rates. The
 259 open vials allowed CO_2 exchange with the atmosphere while preventing downstream wa-
 260 ter leaching. Although these experiments simplified the soil environment considerably,
 261 they provided valuable insights by enabling a direct assessment of the soil-water-air chem-
 262 istry influenced by the mineral dissolution.

263 We conducted model simulations, configuring the numerical parameters to align
 264 with the experimental conditions (Table S1). The sandy soil was characterized by high
 265 acidity (initial pH = 3.55) and substantial organic carbon content (initial OC = 5.5%).
 266 Olivine powder with an average diameter of 20 μm was applied at two distinct rates, equiv-
 267 alent to 1 and 5 kg/m^2 . The soil was constantly moist, and the temperature was fixed
 268 at the experimental value of 22 $^\circ\text{C}$. The initial CO_2 concentration in the soil air was set
 269 at 23 times atmospheric values to reproduce the observed soil respiration flux. Data about
 270 adsorbed species were not provided in the experimental work, so we estimated a CEC
 271 of 10 $\text{cmol}_c/\text{kg}_{\text{soil}}$ with 10% base saturation from literature values for extremely acidic
 272 sandy loam (Weil & Brady, 2016). Different assumptions on the CEC and its base sat-
 273 uration have little quantitative impact on the results (Fig. S4 and S5). The simulations
 274 encompass the three experimental scenarios: control, and low and high olivine applica-
 275 tions.

276 The comparison between simulations and experiments is presented in Fig. 3, which
 277 highlights the Mg accumulation in the vial due to mineral dissolution (a), the soil pH
 278 shifts (b), and the flux of CO_2 from the soil to the atmosphere (c). The simulations closely
 279 align with the experimental findings in all three scenarios. Notably, the Mg accumula-
 280 tions in the vials constrain the weathering rates since there are no Mg losses from the
 281 control volume. As further commented in the discussion (Sec. 4.1), the dissolution fac-
 282 tor F_D in the weathering formula (10) had to be adjusted to values $\mathcal{O}(0.1)$, implying that
 283 Palandri’s formula (10) substantially overestimated the mineral dissolution rate. The pH
 284 shifts show good agreement except in the high-olivine application, suggesting that ex-
 285 perimental soil pH is more buffered than what our numerical simulations reproduce, pos-
 286 sibly due to the assumed CEC (Fig. S5). The CO_2 flux to the atmosphere remains nearly
 287 identical in the control and olivine treatments, implying no CO_2 sequestration in either
 288 experiment or model. This is due to the low pH levels that impede aqueous carbonate
 289 formation (Bertagni & Porporato, 2022; Dietzen & Rosing, 2023). More extended ex-
 290 periments allowing for further olivine dissolution would raise the pH to favorable values
 291 for CO_2 sequestration.

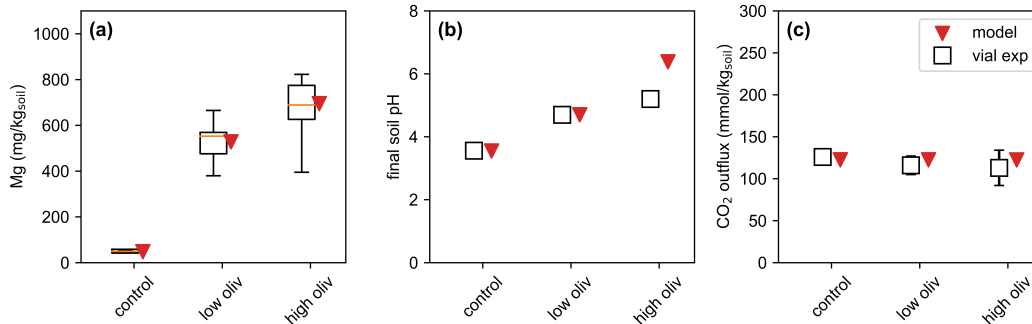


Figure 3. Model-experiment comparison based on the vial experiments by Dietzen et al. (2018). (a) Final Mg accumulation in the soil. For the experimental replicates (10 for each case), the central line represents the median value, the boxes span from the 25th to 75th percentiles, and the whiskers extend from the minimum to the maximum value. (b) Final soil pH (c) Mean cumulative net CO₂ flux to the atmosphere, showing minimal variations across treatments. Error bars indicate experimental standard deviation (SD) – not shown if shorter than the symbol size.

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3.2 Soil Columns with Leaching

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Our second comparison is with experiments conducted by te Pas et al. (2023), featuring small down-flow soil columns of 180 ml polyethylene containers. These columns were equipped with perforated bases to enable water leaching. The experiments thus account for a rudimentary hydrologic cycle, wherein the soil-rock mixture undergoes wet-dry cycles with water added every three days. A further advantage of these nine-week experiments is that they assessed the enhanced weathering potential of five distinct rocks and minerals: forsterite (Mg₂SiO₄), wollastonite (CaSiO₃), anorthite (CaAl₂Si₂O₈), albite (NaAlSi₃O₈), and basalt.

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We conducted model simulations utilizing parameters directly derived from the experiments. The sandy soil had an initial pH of 5.2 and an organic carbon content of 2.1%. Deterministic rainfall events of constant intensity were applied at three-day intervals with deionized water (no alkalinity inputs). The resulting rainfall regime (around 3200 mm/yr) is typical of tropical regions. Rock powder application mirrored the experimental high load of 12.5 kg/m² across all cases, incorporating different particle size distributions and specific surface areas. Albite mineral composition included a 3% of wollastonite. Without data regarding the mineral composition of basalt, we adopted the basalt characterization from Beerling et al. (2020). The temperature was set at 22 °C. Equilibrium-based initial conditions for adsorbed and dissolved species were established based on experimental measurements of total alkaline cation (Ca, Mg, K, Na) quantities. The CEC was fixed at the effective CEC value (3 cmol_c/kg_{soil}) observed at the beginning of the experiments.

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Figure 4 presents the model-experiment comparisons for the total alkalinity release by mineral dissolution (a), the increase in soil pH (b), and the CO₂ captured by the EW applications (c). The total alkalinity release includes alkalinity observed in leaching and soil adsorption (see Fig. S6 for the partitioning between the two phases) and constrains the mineral weathering rates, giving F_D values in the weathering formula (10) consistently below one. The pH shifts show a reasonable agreement, although the numerical simulations do not fully reproduce the increase in pH that is observed experimentally. This is consistent with the experimental increase in soil pH even in the absence of rock application, which the model only partially reproduces. The pH temporal dynamics (Fig. S6)

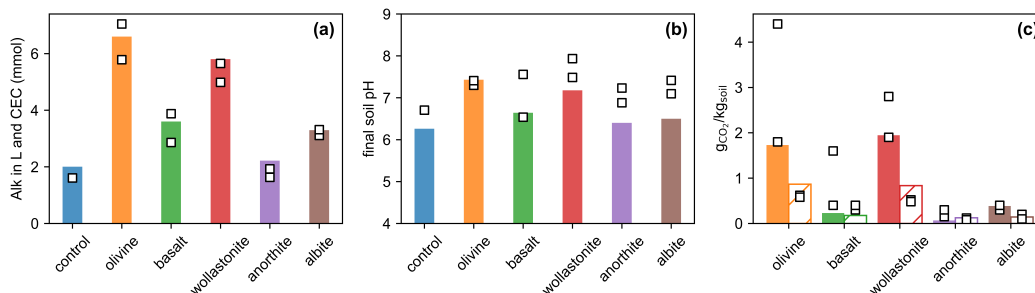


Figure 4. Model-experiment comparison based on the down-flow bottle experiments by te Pas et al. (2023). Square symbols stand for the experimental replicates. (a) Total alkalinity observed in the leaching and on the cation exchange capacity. (b) Final soil pH. (c) Potential (filled bars) and effective (dashed bars) CO₂ sequestration. The difference is due to alkalinity adsorption on the cation-exchange sites.

323 further reveal a model-experiment difference in the first days of the experiment, where
 324 the model does not reproduce the pH experimental drop likely driven by the acidity re-
 325 leased by the cation exchange. Following the experimental work (te Pas et al., 2023), we
 326 quantified the CO₂ sequestration in two ways: one based on the alkalinity liberated through
 327 rock dissolution (potential CO₂ sequestration) and the other accounting for aqueous car-
 328 bonate leaching and additional inorganic carbon stored in the soil (effective CO₂ cap-
 329 ture). The difference between the potential and effective CO₂ sequestration is due to the
 330 alkalinity adsorption on cation-exchange sites, which does not promote aqueous carbon-
 331 ate formation. Consistently with the experiments, the effective CO₂ sequestration is sig-
 332 nificantly lower than the potential CO₂ sequestration across all experiments.

333 3.3 Mesocosms with Vegetation

334 The third and fourth comparisons are with mesocosm experiments (Kelland et al.,
 335 2020; Amann et al., 2020). A distinctive feature of these experiments was the inclusion
 336 of actively growing vegetation, specifically sorghum in Kelland et al. (2020) and wheat
 337 and barley in Amann et al. (2020). Vegetation introduces complexities to soil hydro-
 338 logy and biogeochemistry through water transpiration, nutrient uptake, and CO₂ autotrophic
 339 respiration (Appendix B). Although the representation of the hydrological cycle in these
 340 experiments remained somewhat simplified with periodic (1-7 days) and fixed amounts
 341 of water addition, the dynamic interplay with vegetation growth resulted in notable wa-
 342 ter flux shifts during the growing season. This allowed for an expanded comparative anal-
 343 ysis, including examining hydrological and soil biogeochemical processes.

344 3.3.1 Reactor Columns

345 Kelland et al. (2020) conducted experiments in reactor columns measuring 16 cm
 346 in diameter and 50 cm in depth over 120 days. We ran model simulations based on the
 347 experimentally observed parameters as in the previous comparisons. The soil was clas-
 348 sified as a clay loam with an initial pH of 6.6 and an organic carbon content of 1.2%. The
 349 simulations incorporated a rainfall regime typical of temperate humid and tropical re-
 350 gions (about 2000 mm/yr), with water added every five days. To estimate the poten-
 351 tial evapotranspiration, we numerically recreated the experimental artificial day. This
 352 involved maintaining photosynthetically active radiation ($800 \mu\text{mol photons m}^{-2} \text{s}^{-1}$)
 353 for 18 hr during the initial 60 days and 10 hr for the subsequent 60 days. Daily temper-
 354 atures were computed by temporally averaging the 25 and 17 °C of the artificial day and

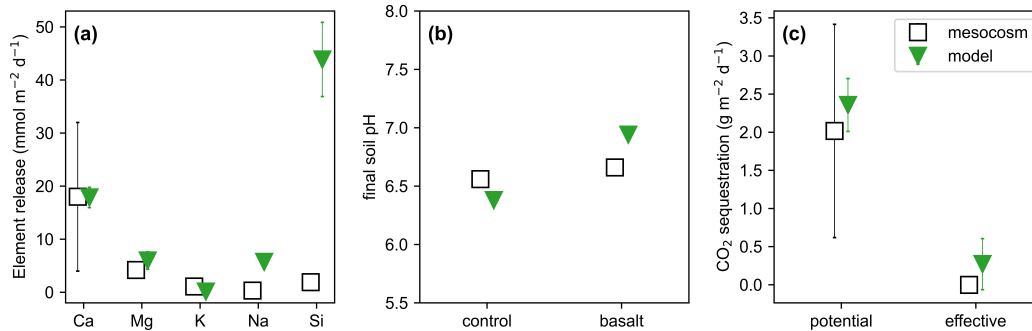


Figure 5. Model-experiment comparison based on the mesocosm experiments by Kelland et al. (2020). (a) Daily averaged elemental release of basalt dissolution per land surface unit, with bars in model results indicating ± 1 SD associated with time variability. (b) Final pH. (c) Potential (alkalinity release) and effective (aqueous carbonate leaching) CO₂ sequestration.

355 night, respectively. We used experimental values for the basalt application (high load
 356 of 10 kg/m²), rock mineral composition, specific surface area, and particle size distribu-
 357 tion. Due to the depth-averaged model framework, we could not reproduce the exper-
 358 imental vertical heterogeneity, with basalt being mixed only in the first 25 cm of the soil
 359 column. The CEC was fixed at the experimental value of 25 cmol_c/kg_{soil}, and the ini-
 360 tial saturation fractions were estimated based on the cation concentrations measured in
 361 the leachate of the untreated experiment.

362 Fig. 5 presents the model-experiment comparison for the elemental release through
 363 basalt dissolution (a), the impact on soil pH (b), and potential and effective CO₂ seques-
 364 tration (c). The model well captures the release of alkaline nutrients Ca, Mg, and K. We
 365 stress, however, that the very fast dissolution of apatite, Ca₅(PO₄)₃(OH), comprising
 366 around 3% of the basalt, could not be numerically reproduced (see the discussion sec-
 367 tion) and has been added at *a posteriori* to the simulation results. The model instead
 368 overestimates the release of Na and Si, suggesting that plants and fungi in the experi-
 369 ments might have driven incongruent dissolution reactions. It might also be that the Si
 370 experimental values are biased low due to underestimation of the Si pool by extraction
 371 with ammonium acetate (Wang et al., 2004). There is also a promising model-experiment
 372 agreement in the Ca, Mg, and Si partitioning among soil, plant, and leachate (Fig. S7).
 373 Moreover, the model effectively reproduces the potential CO₂ sequestration resulting from
 374 alkaline element release, demonstrating substantially higher values in both experiments
 375 and simulations than the effective CO₂ sequestered through aqueous carbonate leach-
 376 ing. The difference is primarily due to CEC adsorption, with plant uptake playing a mi-
 377 nor role (Fig. S7).

378 3.3.2 Rain Barrels

379 Amann et al. (2020) performed experiments in rain barrels measuring 46 cm in di-
 380 ameter and 26 cm in depth over a year. The soil was classified as loamy sand with an
 381 initial pH of 7 and an initial organic carbon content of 1.2%. The simulations mimic a
 382 rainfall regime of 800 mm/yr, with rainfall events distributed every day or week, with
 383 little difference in the results between the two cases. The particle size distributions dif-
 384 ferentiate between coarse and fine olivine (mostly forsterite, Mg₂SiO₄) applications of
 385 22 kg/m², with 25 and 720 μm being the dominant diameter classes, respectively. The
 386 simulations assume that the olivine is mixed across the barrel, while, in the experiments,
 387 olivine was mixed in the top layer of approximately 11 cm. Temperature was varied with

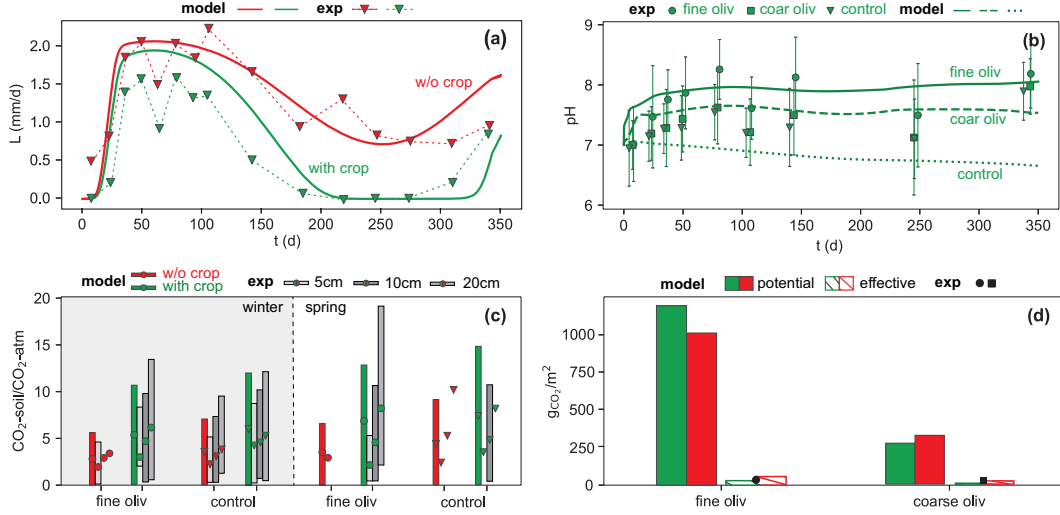


Figure 6. Model-experiment comparison based on the mesocosm experiments by Amann et al. (2020), in the presence (green) and absence (red) of vegetation. (a) Leaching fluxes. Experimental leaching fluxes are from the control experiments. (b) pH dynamics in the presence of vegetation. The reported experimental values are averages between shallow (1.5 cm) and deep (24 cm) measurements. Error bars on experimental values indicate $\pm 1SD$. Fig. S8 shows the equivalent plot in the absence of vegetation. (c) Soil air CO_2 , showing experimental measurements obtained at different depths compared with depth-averaged model results. Bars indicate $\pm 1SD$ associated with time variability (model) and experimental replicates when available. For the numerical results, winter and spring are defined as the experiment’s first and second 100 days, respectively. (d) Potential and effective CO_2 sequestration, with experimental results showcasing averaged effective CO_2 sequestration with and without vegetation.

388 a sinusoidal function across the year, from a minimum of $6^\circ C$ to a maximum of $25^\circ C$.
 389 The CEC was fixed at the experimental value of $8.6 \text{ cmol}_c/\text{kg}_{\text{soil}}$, initially saturated by
 390 86.5% of Ca, 5% of Mg, 5% of K, 3% of Na, and 0.5% of H and Al (Amann et al., 2020).
 391 We ran six simulations for the control case and the coarse and fine olivine applications,
 392 with and without vegetation.

393 Fig. 6 presents the model-experiment comparisons in terms of hydrological balance
 394 (a), pH dynamics (b), soil CO_2 (c), and CO_2 sequestration (d). For this experimen-
 395 tal setup, leaching is a significant proxy for hydrologic partitioning since evapotran-
 396 spiration directly results from the difference between water input and leaching. The sim-
 397 ulations reproduce the leaching seasonal patterns due to the impact of temperature and
 398 vegetation. However, the experiments revealed a slightly different water partitioning due
 399 to rock application altering the soil hydraulic property, which the model cannot repro-
 400 duce (Amann et al., 2020). The soil bulk pH dynamics show reasonable agreement in
 401 the barrels with rock powder applications, with an increase over time due to olivine dis-
 402 solution. Model results and observations do not match for the control barrels, where the
 403 experimental pH shows a considerable increase in soil pH even in the absence of rock pow-
 404 der. Both simulation and experiments show how vegetation tends to reduce the pH by
 405 cation uptake and CO_2 respiration (Fig. 6b and S8). The soil air CO_2 dynamics also show
 406 promising results, with the model reproducing seasonal variations and the influence of
 407 the vegetation in increasing the CO_2 concentration (Fig. 6c).

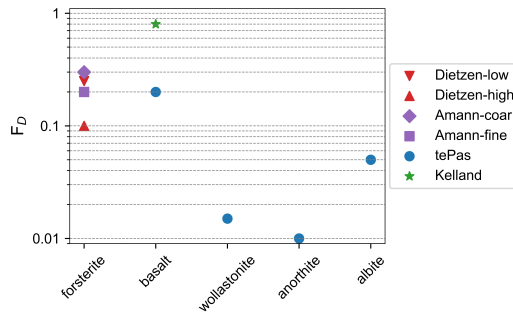


Figure 7. Dissolution factor (F_D) used in the weathering rate equation (10) to reproduce the experimental observations of alkalinity releases.

408 Regarding CO₂ sequestration, we estimated the potential CO₂ capture due to the
 409 Mg released by mineral dissolution and the effective CO₂ capture, here quantified through
 410 the Mg observed in the leached water, as done in the experimental work. Note that the
 411 experimental effective CO₂ capture has been reassessed from Mg leachate data due to
 412 a dimensional inconsistency in the formula (2) reported by Amann et al. (2020). The po-
 413 tential CO₂ capture is much higher than the effective one due to the loss of alkaline cations
 414 to soil adsorption and plant uptake. Interestingly, vegetation has a dual influence on CO₂
 415 removal: plant uptake of alkaline cations reduces the effective CO₂ removal, but plant-
 416 induced soil acidification enhances the mineral dissolution rates, increasing the poten-
 417 tial CO₂ removal. Consequently, our results suggest a trade-off in CO₂ removal efficiency,
 418 with plant-absent scenarios showcasing slower but more efficient removal processes. In
 419 contrast, plant-present scenarios feature faster but less efficient removal processes.

420 4 Discussion

421 The overall favorable agreement between model outcomes and experimental obser-
 422 vations allows us to provide critical insights into the weathering rates. We then identify
 423 and discuss areas requiring further theoretical and experimental exploration.

424 4.1 Weathering Rates

425 The assessment of EW is intricately linked to the precise determination of rock weath-
 426 ering rates, whose parameters are surrounded by considerable uncertainty (Calabrese et
 427 al., 2022). The theoretical formulation used here (Palandri, 2004) is widely regarded as
 428 comprehensive and is commonly applied in EW assessments (Taylor et al., 2016; Beer-
 429 ling et al., 2020; Kantzas et al., 2022; Kanzaki et al., 2022). However, this formulation
 430 is derived from experimental data from stirred reactors without diffusive limitations and
 431 under conditions far from equilibrium. In the complex, multiphase, and porous soil en-
 432 vironment, numerous overlooked biotic and abiotic processes may influence dissolution
 433 rates: i) concentration gradient formation into the aqueous phase near mineral surfaces,
 434 ii) primary and secondary mineral coatings, iii) fungal and bacterial activity, iv) catalyza-
 435 tion or inhibition of the dissolution reactions due to the presence of other chemical species.
 436 While some specific processes are expected to promote dissolution (e.g., biotic activity),
 437 others predominantly impede it (e.g., particle coatings). As a result, the validity of the
 438 formula for EW applications remains an open question.

439 Our model-experiment comparison indicates that Palandri’s formulation requires,
 440 at a minimum, a correction factor for dissolution, F_D in (10), which is consistently be-
 441 low one for most experimental setups and rock types (Fig. 7). This discrepancy under-

442 scores a substantial gap between theoretical and observed dissolution rates, likely due
 443 to the complex abiotic and biotic environmental factors that standard weathering rate
 444 definitions fail to capture. These findings may not be unexpected: similar discrepancies
 445 have frequently been reported in soil rock weathering beyond the context of EW (Brantley,
 446 2003; Jung & Navarre-Sitchler, 2018; Schabernack & Fischer, 2022), as well as in organic
 447 matter decomposition (Davidson & Janssens, 2006), and they align with Palandri’s ob-
 448 servation that *actual equilibration rates are expected to be much slower than those pre-*
 449 *dicted by the selected computer code* (Palandri, 2004). Additionally, while the precise es-
 450 timate of F_D for each specific model-data comparison involves various uncertainties in
 451 process representations and parameter values, these uncertainties are overshadowed by
 452 the much more significant uncertainty in weathering rates, which spans several orders
 453 of magnitude and exerts the most critical control over the entire EW dynamics.

454 The discrepancy between theoretical expectations and observations carries signifi-
 455 cant implications. Weathering rates are the main control of the carbon sequestration
 456 process. Unless field weathering rates are substantially higher than those observed in small-
 457 scale experiments, our results suggest that previous model-based EW assessments - con-
 458 sidering $F_D \geq 1$ due to biotic processes (Beerling et al., 2020; Kantzas et al., 2022) -
 459 may overestimate the potential of EW for CO₂ removal by orders of magnitude. To ac-
 460 curately assess the feasibility and effectiveness of EW in climate mitigation, narrowing
 461 down uncertainties in weathering rates will be essential. Both model advances and ex-
 462 tensive data collection will be required to gain mechanistic insights into F_D and shift mod-
 463 els like SMEW from needing an observation-based calibration of weathering rates to mod-
 464 els with inherent predictive capabilities. Emergent field and laboratory data collection
 465 will be critical in constraining the relationships between weathering rates and specific
 466 mineral types, soil and crop characteristics, and climates. Model improvements will need
 467 to systematically incorporate the above-mentioned soil biotic and abiotic processes that
 468 can influence weathering rates, including microbial activity and mineral surface reactiv-
 469 ity (Schabernack & Fischer, 2022).

470 4.2 Limitations and Outlook

471 The model-experiment comparison presented in this study is only a first step to-
 472 ward having robust assessments of EW models to reproduce observations across scales.
 473 Many advances will be needed from both modeling and observational perspectives.

474 From a modeling point of view, the model presented here accounts for the primary
 475 variables of interest to assess the fate of the alkaline cations released by the EW appli-
 476 cations and their corresponding inorganic carbon sequestration potential. In addition
 477 to model advances needed to gain mechanistic insights into weathering rates, extensions
 478 could include feedback that the rock powder application may have on some soil phys-
 479 ical and biotic processes. The different leaching fluxes observed in the experiments by
 480 Amann et al. (2020) suggest that the rock powder impacts the soil texture and hydraulic
 481 conductivity, hence the soil water partitioning. Specific experiments evaluating the tem-
 482 poral evolution of soil physical properties are needed to incorporate such feedback in mod-
 483 els, even though some theoretical estimates may be derived based on soil physics mod-
 484 els (Jury & Horton, 2004). Furthermore, rock applications may influence biotic activ-
 485 ity and the organic carbon balance, with potentially detrimental effects in tropical soils
 486 and peatlands (Klemme et al., 2022). Modeling advances could also include mass bal-
 487 ances for heavy metal accumulations, such as nickel and copper, which are significant
 488 concerns in the context of EW applications (Haque et al., 2020; Dupla et al., 2023).

489 Compared to the more popular reactive transport models, SMEW is more parsim-
 490 onious, not accounting for soil vertical heterogeneity and including fewer chemical species.
 491 This simplicity comes at the cost of spatial information but at the advantage of acces-
 492 sibility and a more focused examination of temporal dynamics. Interestingly, our model

493 does not necessitate a semi-empirical pH buffer function often employed in EW simu-
494 lations with reactive transport models to avoid unrealistic spikes in soil pH. This is in-
495 triguating since our buffering mechanisms incorporate only carbonate chemistry and cation
496 adsorption while neglecting others like organic alkalinity. On the contrary, our model en-
497 counters limitations in reproducing the rapid dissolution of certain minerals and mate-
498 rials (e.g., $\text{Ca}(\text{OH})_2$) due to pronounced spikes in alkalinity that hinder the numerical
499 convergence of the implicit system (Appendix A). Looking ahead, a promising avenue
500 involves integrating our model results with both reactive transport models and obser-
501 vational data to gain comprehensive insights into soil EW dynamics.

502 From an experimental standpoint, it is worth acknowledging the temporal constraint
503 within the available datasets (Table S1). Specifically, only one of the experimental datasets
504 used within this study spans a complete year (Amann et al., 2020), while others have
505 a relatively shorter duration of a few months. Given the potential yearly timescales as-
506 sociated with the dissolution of EW rock powder, extrapolating results becomes chal-
507 lenging, especially considering that weathering rates may decrease over the years (Fig. S1).
508 Additionally, most experiments relied on elevated rock loadings (i.e., $\geq 10 \text{ kg/m}^2$) to
509 enhance signals within the short experimental time frame, although such loadings may
510 not be realistic for practical applications. There is also an opportunity to explore the
511 influence of realistic stochastic rainfall regimes, often absent in current experimental se-
512 tups.

513 Integrating field observations of large-scale EW applications will hopefully address
514 some of these temporal and loading limitations, offering insights into the alignment be-
515 tween model results, small-scale experiments, and the practical considerations of large-
516 scale field trials. Optimizing the spatial and temporal frequency of data collection will
517 be crucial to planning feasible field campaigns while preserving quantitative information
518 on soil heterogeneity and temporal dynamics caused by seasonal and daily fluctuations
519 in hydrological and biogeochemical quantities. Priority should be given to measuring the
520 partitioning of released alkaline cations among leaching fluxes, plant uptake, and soil ad-
521 sorption. This serves a dual purpose: (i) quantifying weathering rates and (ii) understand-
522 ing the discrepancy between effective and potential CO_2 sequestration. Additionally, since
523 the movement of alkaline cations extends into deeper soil layers and stream networks,
524 conducting coupled measurements within connected streams can help quantify the ac-
525 tual travel time and flux of the weathering products. Lastly, although SMEW is pars-
526 monious with respect to the complexity of hydrological and biogeochemical processes in
527 soils, it still consists of multiple state variables, parameters, and highly nonlinear inter-
528 actions. With the increasing availability of data from laboratory and field experiments,
529 Bayesian approaches with information criteria metrics can be a valuable approach to solv-
530 ing inverse problems, simultaneously estimating parameters and their uncertainty while
531 also accounting for model complexity.

532 5 Conclusions

533 While enhanced weathering (EW) holds great promise as a negative emission strat-
534 egy, thanks to its significant CDR potential, low technological prerequisites, and valu-
535 able co-benefits, no model has been shown to reproduce EW observations at scale. This
536 deficit restricts our ability to make accurate quantitative predictions for assessments of
537 CDR via EW. In this study, we took a benchmark step in this direction, developing a
538 relatively accessible ecohydrological and biogeochemical model whose results could be
539 meticulously compared with four distinct experimental datasets of different complexity.

540 The model-experiment comparison demonstrates an overall favorable agreement
541 for the primary variables of interest, including water partitioning, alkalinity release, pH
542 dynamics, and CO_2 sequestration. The comparison also demonstrates that weathering
543 rates are lower than traditionally assumed by one or two orders of magnitudes and high-

lights further research directions to improve our understanding and quantitative predictive power for EW as a NET. Finally, while representing EW dynamics within the soil's upper layers is crucial, EW negative emission potential is linked to the fate of rock dissolution products from the field to the ocean (Hartmann et al., 2013; Calabrese et al., 2022; Bertagni et al., 2024), a journey yet to be fully disclosed.

Appendix A Implicit System of Equilibrium Equations

We here report the implicit system of equilibrium equations solved under the quasi-steady approximation jointly with the system of ODEs (1)-(8). These equations are all coupled and quantify how total quantities within the soil control volume of depth Z are distributed among the different soil phases. Specifically, alkaline cations (X_{tot}) are distributed between dissolved and adsorbed phases, inorganic carbon (IC_{tot}) is distributed between aqueous and air phases, and aluminum (Al_{tot}) exists dissolved in water, adsorbed to the soil matrix or in complexes with organic or inorganic matter. In formula

$$X_{\text{tot}} = nZs[X] + f_X \text{CEC}/n_X, \quad (\text{A1})$$

$$\text{IC}_{\text{tot}} = nZs[\text{DIC}] + nZ(1-s)[\text{CO}_2]_a, \quad (\text{A2})$$

$$\text{Al}_{\text{tot}} = nZs[\text{Al}]_{\text{mob}} + f_{\text{Al}} \text{CEC}/3 + \text{Al}_{\text{imm}}, \quad (\text{A3})$$

where n_X is the cation valence and CEC is the cation exchange capacity. The latter indicates the moles of dissolved cations that can be adsorbed on soil colloids due to their negatively charged surface (Weil & Brady, 2016).

Cation Partitioning and Soil Adsorption. The master variable connecting alkaline cations and carbonate system is alkalinity (Alk). Expressed in terms of species that are conservative to changes in pH, temperature, and pressure (Wolf-Gladrow et al., 2007; Bertagni & Porporato, 2022), alkalinity is

$$[\text{Alk}] = 2[\text{Ca}^{2+}] + 2[\text{Mg}^{2+}] + [\text{K}^{2+}] + [\text{Na}^{2+}] - [\text{An}], \quad (\text{A4})$$

where $[\text{An}]$ indicates the cumulative concentration of the anions of the strong acids. Quantifying the dissolved cations in the soil solution requires assessing the cation partitioning between the dissolved and adsorbed phases. This is done using the Gaines-Thomas convention (Bleam, 2017). Specifically, five equations are used to describe the binary exchange of Ca^{2+} with Al^{3+} , Mg^{2+} , Na^+ , K^+ , and H^+ :

$$\frac{f_{\text{Ca}}^3}{f_{\text{Al}}^2} = K_{\text{Ca-Al}} \frac{[\text{Ca}^{2+}]^3}{[\text{Al}^{3+}]^2}, \quad \frac{f_{\text{Ca}}}{f_{\text{Mg}}} = K_{\text{Ca-Mg}} \frac{[\text{Ca}^{2+}]}{[\text{Mg}^{2+}]}, \quad \frac{f_{\text{Ca}}}{f_{\text{K}}^2} = K_{\text{Ca-K}} \frac{[\text{Ca}^{2+}]}{[\text{K}^+]^2}, \quad (\text{A5})$$

where the exchange equations for Ca-Na and Ca-H are equivalent to Ca-K. The soil-dependent cation exchange constants can be evaluated with coupled measurements of adsorbed and dissolved species or can be evaluated after the extensive dataset of Vries and Posch (2003). The sum of all exchangeable fractions (f) is equal to unity, namely

$$f_{\text{Ca}} + f_{\text{Al}} + f_{\text{Mg}} + f_{\text{Na}} + f_{\text{K}} + f_{\text{H}} = 1. \quad (\text{A6})$$

Air-Water Carbonate System. In the soil solutions, and more generally in natural waters, the alkalinity charge difference expressed in (A4) is balanced by the aqueous carbonate system

$$[\text{Alk}] = [\text{HCO}_3^-] + 2[\text{CO}_3^{2-}] + [\text{OH}^-] - [\text{H}^+], \quad (\text{A7})$$

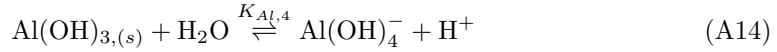
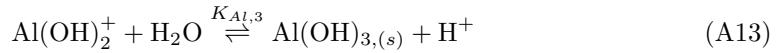
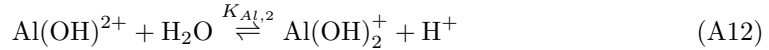
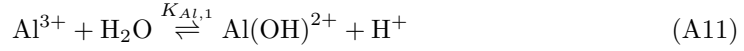
$$[\text{DIC}] = [\text{CO}_2]_w + [\text{HCO}_3^-] + [\text{CO}_3^{2-}] \quad (\text{A8})$$

$$[\text{HCO}_3^-] = K_1[\text{CO}_2]_w/[\text{H}^+], \quad [\text{CO}_3^{2-}] = K_1K_2[\text{CO}_2]_w/[\text{H}^+]^2, \quad (\text{A9})$$

$$[\text{CO}_2]_a = K_{\text{H}}[\text{CO}_2]_w, \quad [\text{OH}^-] = [\text{H}^+]/K_w, \quad (\text{A10})$$

553 where K_1 and K_2 are the first and second carbonic acid dissociation constants. K_w is
 554 the water dissociation constant. K_H is Henry's constant for CO_2 solubility. All these con-
 555 stants and their temperature dependence are evaluated after Stumm and Morgan (1996).
 556 The combination of Eqs. (A4) and (A7) summarize EW goal of increasing alkalinity by
 557 mineral dissolution to promote aqueous carbonate formation. The efficiency of this pro-
 558 cess varies as a function of the water chemistry (Bertagni & Porporato, 2022). Formal
 559 extension to the alkalinity definition (A7) could include aluminum, which plays a buffer
 560 role in acidic conditions, and organic alkalinity. Other weak acids and bases have been
 561 shown to play a negligible role in the soil solution (Bertagni & Porporato, 2022).

Aluminum Speciation. Aluminum chemistry is complex and strongly influenced
 by water pH (Weil & Brady, 2016; Nordstrom & May, 2020). Aluminum in aqueous sys-
 tems speciates into five main monomeric species, following the reactions



where the constants K_{Al} are evaluated after Weil and Brady (2016). In highly acidic ($\text{pH} < 4.5$) and highly alkaline ($\text{pH} > 7$) conditions, aluminum solubility is enhanced, and the dominant species are dissolved Al^{3+} and $\text{Al}(\text{OH})_4^-$, respectively. By contrast, at intermediate pH values ($5 < \text{pH} < 7$), Al is present in less mobile forms, such as the hydroxy aluminum ions $\text{Al}(\text{OH})^{2+}$ and $\text{Al}(\text{OH})_2^+$, which typically form complexes with organic matter and other soil elements, as well as the solid mineral gibbsite $\text{Al}(\text{OH})_3$. We hence discriminate into mobile ($[\text{Al}]_{\text{mob}}$) and immobile ($[\text{Al}]_{\text{imm}}$) aluminum pools following

$$[\text{Al}]_{\text{mob}} = [\text{Al}^{3+}] + [\text{Al}(\text{OH})_4^-], \quad (\text{A15})$$

$$[\text{Al}]_{\text{imm}} = ([\text{Al}(\text{OH})^{2+}] + [\text{Al}(\text{OH})_2^+] + [\text{Al}(\text{OH})_3])nZ_r s. \quad (\text{A16})$$

562 We then consider that only the mobile Al can be lost through leaching events; see eq. (8).

563 Appendix B Plants dynamics and their role in EW

In SMEW, plants influence enhanced weathering dynamics by impacting soil hydrological and biogeochemical balances. Plant roots transpire water, actively and passively uptake nutrients, and release inorganic carbon (autotrophic respiration). Growing vegetation (V) can be dynamically modeled through a classical logistic equation

$$\frac{dV}{dt} = \alpha_V V(k_V - V), \quad (\text{B1})$$

564 where k_V is the carrying capacity per unit area, dependent on plant and ecosystem types,
 565 and α_V is the plant growth rate. The growth rate can be estimated based on the time
 566 (t_V) required for plants to progress from seedling to maturity through $\alpha_V \approx 6/t_V$. Sim-
 567 ulated plant-mediated processes are then scaled with the normalized vegetation variable
 568 $\hat{V} = V/k_V$, defined between 0 and 1. These processes include: i) plant transpiration
 569 (T), modeled as a soil moisture function (Laio et al., 2001); ii) plant passive uptake, as-
 570 sumed to be directly proportional to the transpiration rate (Cipolla et al., 2021a); iii)
 571 autotrophic respiration (RESP_a), estimated to be equivalent to heterotrophic respira-
 572 tion when plants are fully grown (Bond-Lamberty et al., 2004); iv) active uptake, which
 573 occurs during plant growth when passive uptake alone cannot meet the nutrient demands
 574 for growth (see below). Noteworthy, both plant-mediated nutrient uptake and inorganic
 575 carbon release processes contribute to soil acidification.

In addition to the nutrient uptake through the transpiration stream (passive uptake), plants can also rely on more complex and energetically expensive physiological processes (active uptake) when the passive uptake is insufficient to meet the nutrient demand (DEM). The active uptake then counts on a diffusion flux from the bulk of the solution to the plant roots (Porporato, D’Odorico, et al., 2003; Grathwohl, 1998; Porporato & Yin, 2022). Here we propose a new modeling framework for the plant active uptake wherein the diffusive flux is quantified by the root surface area (i.e., the root area index, RAI), the element diffusivity in water (D_w), and the concentration gradient between the root surface and the solution bulk. Assuming a null element concentration on the root surface and taking calcium (Ca) as an example, the concentration gradient is $[Ca]/\ell$, where ℓ is the typical distance traveled from bulk to root. The latter can be quantified as $\ell = \sqrt{d_r Z / (\hat{V} \cdot \text{RAI})}$, assuming parallel cylindrical roots of average diameter d_r uniformly distributed over the depth Z (Manzoni et al., 2013) and considering that the root surface area scales with the vegetation stage. Active uptake for Ca can then be expressed as

$$\text{UP}_{\text{Ca}} = \begin{cases} 0 & \text{if } [Ca]T \geq \text{DEM}_{\text{Ca}} \\ \min\left(\hat{V} \cdot \text{RAI} \cdot D_w \frac{[Ca]}{\ell}, \text{DEM}_{\text{Ca}} - T[Ca]\right) & \text{if } [Ca]T < \text{DEM}_{\text{Ca}} \end{cases} \quad (\text{B2})$$

576 DEM_{Ca} defines the calcium required for the plant’s new biomass development ($\text{DEM}_{\text{Ca}} =$
577 $\xi_{\text{Ca}} dV/dt$), with ξ_{Ca} being a plant-dependent coefficient specifying moles of Ca per biomass
578 unit. Similar equations apply to other essential plant nutrients, including Mg, K, and
579 Si.

580 **Data and Software Statement** The numerical codes for SMEW (Python), the
581 Jupyter Notebooks for the model-experiment comparisons, and all numerical data pro-
582 duced within this manuscript are available on GitHub ([https://github.com/MatteoBertagni/](https://github.com/MatteoBertagni/SMEW)
583 [SMEW](https://github.com/MatteoBertagni/SMEW)) and on Zenodo (upon publication). The experimental data come from previous
584 works, as acknowledged in the manuscript.

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