



## Abstract

Enhanced weathering (EW) is a promising strategy to remove atmospheric CO<sub>2</sub> by amending agricultural and forestry soils with ground silicate materials. However, the current model-based assessments of EW potential face uncertainties stemming from the intricate interplay among soil physical, chemical, and biotic processes, compounded by the absence of a detailed model-data comparison, mostly due to the limited availability of comprehensive data. Here, we address this critical gap by advancing and validating an ecohydrological and biogeochemical model for EW dynamics in soils. We conduct a hierarchical validation in which model results are critically compared to four experimental datasets of increasing complexity, from simple closed incubation systems to open mesocosm experiments. The comparison demonstrates the model ability to capture the dynamics of primary variables, including rock alkalinity release and CO<sub>2</sub> sequestration. The comparison also reveals that weathering rates are consistently lower than traditionally assumed by up to two orders of magnitude. We finally discuss 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 enriching cropland and forest soils with finely ground silicate rocks, which sequester atmospheric CO<sub>2</sub> 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 rate than conventionally assumed, offering crucial insights for a comprehensive understanding of EW 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> removal per year (Taylor et al., 2016; Strefler et al., 2018; Beerling et al., 2020; Baek et al., 2023).

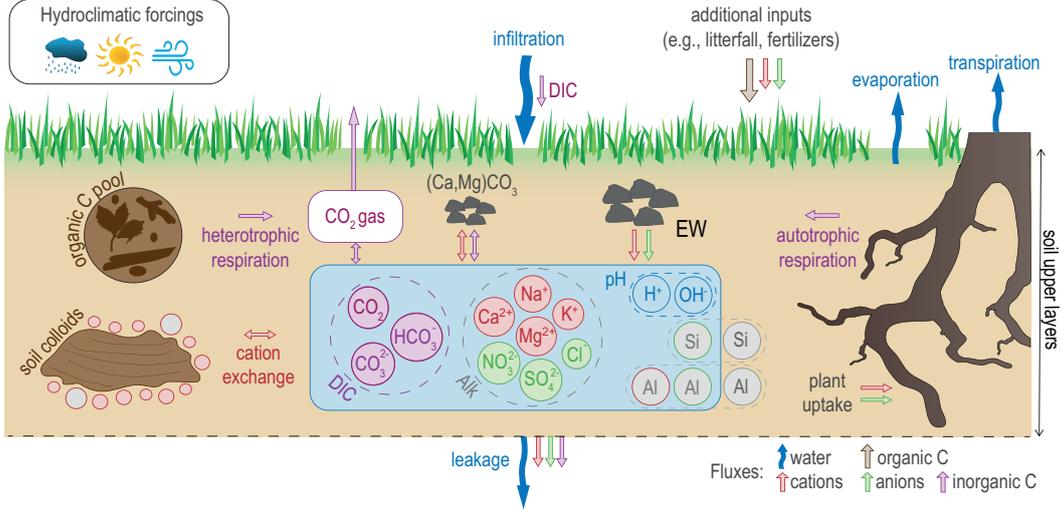
Despite these promising estimates, EW assessments face significant uncertainties rooted in the complex interplay between hydrological and biogeochemical processes across scales (Calabrese et al., 2022). Weathering rates exhibit considerable variability, spanning orders of magnitudes due to temporal and spatial heterogeneity in hydroclimatic drivers and soil processes (Amann et al., 2020; Calabrese & Porporato, 2020; Deng et

68 al., 2022). This variability complicates efforts to reconcile theoretical expectations with  
69 laboratory observations, even for the same rock type (Renforth et al., 2015; Amann et  
70 al., 2020; Buckingham et al., 2022). Field trials are in their nascent stages, and moni-  
71 toring, reporting, and verifying (MRV) carbon dioxide removal in open, heterogeneous,  
72 and multiphase soil systems face inherent challenges (Clarkson et al., 2023), although  
73 recent efforts have started to address them (Amann & Hartmann, 2022; Reershemius et  
74 al., 2023; Knapp et al., 2023). Specifically, soil-based mass balance approaches, initially  
75 used for natural chemical weathering but modified to reduce analytical error, could be  
76 a promising option to resolve the small signal-to-noise ratios in EW mineral cation de-  
77 pletion (Reershemius et al., 2023). When EW is applied to significant portions of a wa-  
78 tershed area, stream water chemistry analyses may also be beneficial to understand the  
79 transport of the mineral dissolution products (Larkin et al., 2022), as in the case of agri-  
80 cultural liming (Hamilton et al., 2007). Quantifying this transport from the field to CO<sub>2</sub>  
81 storage locations, such as deep aquifers or the oceans, is a crucial and yet largely unex-  
82 plored EW aspect (Hartmann et al., 2013; Zhang et al., 2022; Bertagni & Porporato, 2022;  
83 Calabrese et al., 2022).

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

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

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



**Figure 1.** Sketch of the biogeochemical and ecohydrological processes represented in the model.

## 2 EW Model for the Upper Soil Layers

This model builds upon prior work on EW in the topsoil 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 like mixing the ground rock 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.

### 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 our modeling framework, 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., 2001b; 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 ( $Z$ )

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

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

## 147 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  $\text{CO}_2$  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

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

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

## 159 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  $\text{CO}_2$  removal efficiency (Hartmann et al., 2013; Bertagni & Porporato, 2022). The main components of soil inorganic carbon include  $\text{CO}_2$  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:

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

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

160 where  $[\cdot]$  indicates concentration.  $RESP_{h+a}$  is the sum of heterotrophic and autotrophic  
 161 respiration, respectively. Autotrophic respiration ( $RESP_a$ ), namely the release of  $CO_2$   
 162 gas by plant roots, is estimated to scale with vegetation (Appendix B) and to be equiv-  
 163 alent to heterotrophic respiration (Sec. 2.2) when plants are fully grown (Bond-Lamberty  
 164 et al., 2004). Another minor source of inorganic carbon in the soil is the DIC in infil-  
 165 trating water ( $I_w = R - Q$ ). IC can exit the control volume as aqueous DIC through  
 166 leaching ( $L \cdot [DIC]$ ) or as gaseous  $CO_2$  to the atmosphere through diffusive or advec-  
 167 tive fluxes ( $F_{ADV+DIFF}$ ) (Cipolla et al., 2021a). The term  $W_{(Ca, Mg)CO_3}$  accounts for the  
 168 dissolution ( $> 0$ ) or precipitation ( $< 0$ ) of calcium and magnesium carbonates, mod-  
 169 eled following (Kirk et al., 2015). The redistribution of IC between soil air  $CO_2$  and aque-  
 170 ous carbonates follows equilibrium assumptions (Appendix A).

## 171 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

$$\frac{dX_{tot}}{dt} = I_X + EW_X + W_{XCO_3} - (L + T)[X] - UP_X. \quad (5)$$

172  $I_X$  accounts for background cation inputs like litterfall decomposition, fertilizer addition,  
 173 and background weathering processes.  $EW_X$  denotes the cation release by the EW ap-  
 174 plication, and  $W_{XCO_3}$  is the release from the weathering of Ca or Mg carbonate. The  
 175 term  $(L+T)[X]$  characterizes the outflow resulting from leaching and plant passive up-  
 176 take, while  $UP_X$  pertains to active plant uptake during growth (Appendix B). Given the  
 177 total cation amount in the control volume, the partitioning between adsorbed and aque-  
 178 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.,  $Cl^-$ ,  $NO_3^{2-}$ ,  $SO_4^{2-}$ ). Conveniently, we do not need to discrim-  
 inate between the various anions of the strong acids because i) EW aims to increase cation  
 concentrations, ii) anion adsorption 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

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

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

## 181 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). Soil Si chemistry is rather com-  
 plex, 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  $CO_2$   
 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

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

182 where  $I_{\text{Si}}$  represents background Si inputs,  $\text{EW}_{\text{Si}}$  accounts for Si released through EW  
 183 applications,  $(L + T)[\text{Si}]$  signifies Si outflow due to leaching and passive plant uptake,  
 184 and  $\text{UP}_{\text{Si}}$  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

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

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

## 190 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 ( $\text{SA}_i$ ), and can be expressed as

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

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

For the weathering rates, we follow previous EW modeling efforts (Taylor et al.,  
 2016; Beerling et al., 2020; Kantzas et al., 2022) and use the semi-empirical formula by  
 Palandri (2004). This seminal formulation, originally developed in the work of Lasaga  
 (1984), is based on dissolution experiments in stirred reactors without diffusive limita-  
 tions and under conditions far from equilibrium. The formula accounts for the most well-  
 studied mechanisms of mineral dissolution, driven by the water species  $\text{H}_2\text{O}$ ,  $\text{H}^+$ , and  
 $\text{OH}^-$ . As previously implemented by Cipolla et al. (2021a), we also consider the depen-  
 dence of mineral dissolution rates on the relative soil moisture value ( $s$ ) to account for  
 the wet portion of the mineral surface that can actually undergo dissolution. The for-  
 mula for the weathering rate of each mineral can thus be expressed as:

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

197 where  $j$  is the individual weathering agent ( $\text{H}_2\text{O}$ ,  $\text{H}^+$ ,  $\text{OH}^-$ ) and  $a_j$  are the agent  
 198 activities (here approximated as concentrations).  $k_{i,j}(\Theta)$  are the mineral- and agent-dependent  
 199 rates accounting for temperature ( $\Theta$ ) effects, and  $n_{i,j}$  are the reaction order constants

(Palandri, 2004).  $\Omega_i$  is the mineral saturation index (Morel et al., 1993). The coefficients  $p_{i,j}$  and  $q_{i,j}$  have been quantified only for very few minerals and agents and are approximated to unity (Palandri, 2004).  $F_D$  is a dissolution factor that we will quantify based on experimental observations. As later discussed (Sec. 4.1), the observation-driven quantification of  $F_D$  is crucial to assess actual weathering rates and the validity of Palandri’s formulation in the soil environment.

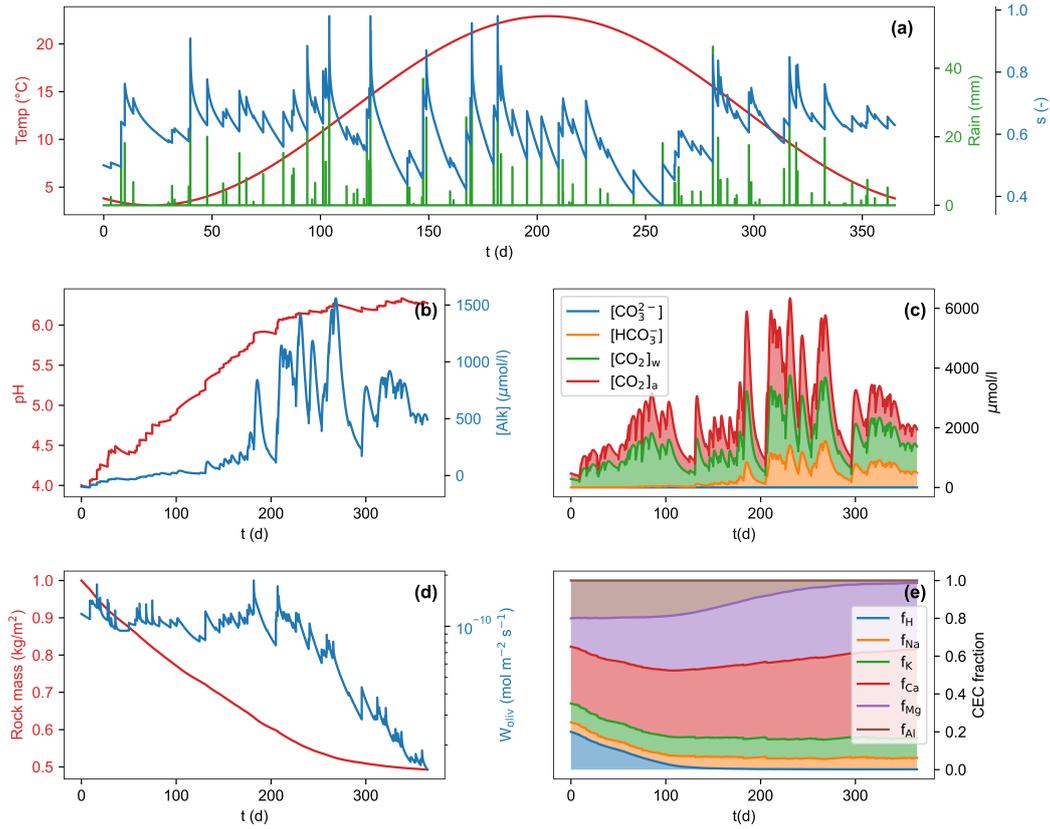
## 2.7 Model Setup and Simulation Example

The EW model requires a characterization of the hydroclimatic forcings, the EW material and application rate, the plant, and the soil biophysical properties. Rainfall and temperature are key hydroclimatic forcings that impact virtually all model components. Wind speed and albedo play a role in influencing potential evapotranspiration (Allen et al., 1998). For EW application, details include the amount and mineral composition of the applied rock, particle size distribution, and specific surface area. Plant characterization involves parameters like carrying capacity, growth rate, and root area index, measured or derived from the literature (Appendix B). Soil characterization requires details on soil texture, initial organic carbon content and pH, and inorganic chemistry. Notably, assuming a quasi-steady state equilibrium of the soil chemistry allows constraining the air-water carbonate system using a single quantity (e.g., the  $\text{CO}_2$  partial pressure) in addition to the soil pH (Stumm & Morgan, 1996). For the biogeochemistry of the major ions, minimum initial data requires either the adsorbed fractions on the cation exchange capacity (CEC), the concentrations in the soil solution, or the total quantities per unit of soil. The others are determined through the Gaines-Thomas convention (Appendix A). In the absence of specific data, background elemental input fluxes, e.g.,  $I_X$  in eq. (5), can be defined to balance background losses, e.g.,  $[x]_{\text{initial}}(\bar{T} + \bar{L})$  where the bar denotes temporal averaging. This approach ensures that the initial condition represents a long-term average state of the soil that is reestablished whenever the EW application is absent or concluded.

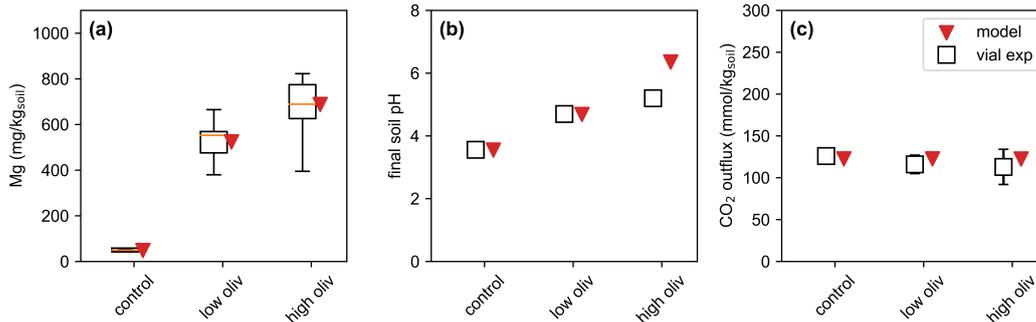
Fig. 2 shows an example of a 1-year simulation for a  $1 \text{ kg/m}^2$  (10 ton/ha) EW application with forsterite ( $\text{Mg}_2\text{SiO}_4$ ) in a temperate humid climate (10-year results are reported in Fig. S1). The mean temperature is  $13^\circ\text{C}$ , and the yearly cumulative rainfall is 1200 mm. The simulated soil is an organic-rich (initial OC is 5%) and acidic (initial pH is 4) loam. Results show how the low soil pH favors the mineral dissolution rate but impedes the formation of bicarbonate in the soil solution (Bertagni & Porporato, 2022; Dietzen & Rosing, 2023). The trend reverses when the pH gets around 6, with slower dissolution rates and bicarbonate formation. The CEC base saturation increases as the pH rises, and  $\text{Mg}^{2+}$  and  $\text{Ca}^+$  replace the acid ions  $\text{H}^+$  and  $\text{Al}^{3+}$ , showing the potential of EW for liming purposes.

## 3 Comparison with Experiments

This section compares our model outcomes with available experimental data. Specifically, we use four different experimental datasets derived from experiments conducted at varying levels of complexity: i) small-scale vials open to the atmosphere and with moist acidic soils (Dietzen et al., 2018); ii) small down-flow soil columns open to the atmosphere and water leaching (te Pas et al., 2023); iii) two more complete mesocosm experiments incorporating growing vegetation (Kelland et al., 2020; Amann et al., 2020). The comparison with these different experimental setups gives the advantages of compartmental investigations of the model performance and a broad examination of EW dynamics under different environmental forcings. Because our model is spatially lumped and designed for the upper soil layers, we could not include in the analysis experiments focusing on EW in vertically deep and heterogeneous soil cores (Renforth et al., 2015; Vienne et al., 2022; Buckingham et al., 2022).



**Figure 2.** Example of model output for an EW application ( $1 \text{ kg/m}^2$ ) with forsterite ( $\text{Mg}_2\text{SiO}_4$ ) in a temperate humid climate. (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. See Fig. S1 for the results extended to 10 years.



**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 CO<sub>2</sub> 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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### 3.1 Acidic Soil in Vials

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Our first comparison involves the experiments by Dietzen et al. (2018), which entailed a three-month soil incubation study to assess the weathering of olivine (mainly composed of forsterite, Mg<sub>2</sub>SiO<sub>4</sub>) and its impact on available Mg levels, pH, and soil CO<sub>2</sub> flux. The experiments employed 110 ml open vials filled with soil that remained consistently moist throughout the study. Olivine was added at varying application rates. The open vials allowed CO<sub>2</sub> exchange with the atmosphere while preventing downstream water leaching. Although these experiments simplified the soil environment considerably, they provided valuable insights by enabling a direct assessment of the soil-water-air chemistry influenced by the mineral dissolution.

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We conducted model simulations, configuring the numerical parameters to align with the experimental conditions. The sandy soil was characterized by high acidity (initial pH = 3.55) and substantial organic carbon content (initial OC = 5.5%). Olivine powder with an average diameter of 20 μm was applied at two distinct rates, equivalent to 1 and 5 kg/m<sup>2</sup>. The soil was constantly moist, and the temperature was fixed at the experimental value of 22 °C. The initial CO<sub>2</sub> concentration in the soil air was set at 23 times atmospheric values to reproduce the observed soil respiration flux. Data about adsorbed species were not provided in the experimental work, so we estimated a CEC of 10 cmol<sub>c</sub>/kg<sub>soil</sub> with 10% base saturation from literature values for extremely acidic sandy loam (Weil & Brady, 2016). The simulations encompass the three experimental scenarios: control and low and high olivine applications.

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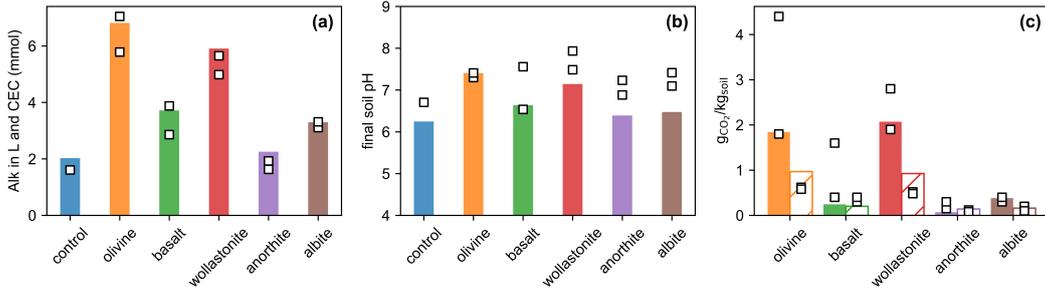
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The comparison between simulations and experiments is presented in Fig. 3, which highlights the Mg accumulation in the vial due to mineral dissolution (a), the soil pH shifts (b), and the flux of CO<sub>2</sub> from the soil to the atmosphere (c). The simulations closely align with the experimental findings in all three scenarios. Notably, the Mg accumulations in the vials constrain the weathering rates since there are no Mg losses from the control volume. As further commented in the discussion (Sec. 4.1), the dissolution factor  $F_D$  in the weathering formula (10) had to be adjusted to values  $\mathcal{O}(0.1)$ , implying that Palandri's formula (10) overestimated the mineral dissolution rate. The pH shifts show good agreement except in the high-olivine application, suggesting that experimental soil pH is more buffered than what our numerical simulations reproduce. The CO<sub>2</sub> flux to



**Figure 4.** Model-experiment comparison based on the down-flow bottle experiments by te Pas et al. (2023). Square symbols stay 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) CO<sub>2</sub> sequestration.

281 the atmosphere remains nearly identical in the control and olivine treatments, imply-  
 282 ing no CO<sub>2</sub> sequestration in either experiment or model. This is due to the low pH lev-  
 283 els that impede aqueous carbonate formation (Bertagni & Porporato, 2022; Dietzen &  
 284 Rosing, 2023). More extended experiments allowing for further olivine dissolution would  
 285 raise the pH to favorable values for CO<sub>2</sub> sequestration.

### 286 3.2 Soil Columns with Leaching

287 Our second comparison is with experiments conducted by te Pas et al. (2023), fea-  
 288 turing small down-flow soil columns of 180 ml polyethylene containers. These columns  
 289 were equipped with perforated bases to enable water leaching. The experiments thus ac-  
 290 count for a rudimentary hydrologic cycle, wherein the soil-rock mixture undergoes wet-  
 291 dry cycles with water added every three days. A further advantage of these nine-week  
 292 experiments is that they assessed the enhanced weathering potential of five distinct rocks  
 293 and minerals: forsterite (Mg<sub>2</sub>SiO<sub>4</sub>), wollastonite (CaSiO<sub>3</sub>), anorthite (CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>), al-  
 294 bite (NaAlSi<sub>3</sub>O<sub>8</sub>), and basalt.

295 We conducted model simulations utilizing parameters directly derived from the ex-  
 296 periments. The sandy soil had an initial pH of 5.2 and an organic carbon content of 2.1%.  
 297 Deterministic rainfall events of constant intensity were applied at three-day intervals.  
 298 The resulting rainfall regime (around 3200 mm/yr) is typical of tropical regions. Rock  
 299 powder application mirrored the experimental high load of 12.5 kg/m<sup>2</sup> across all cases,  
 300 incorporating different particle size distributions and specific surface areas. Albite min-  
 301 eral composition included a 3% of wollastonite. Without data regarding the mineral com-  
 302 position of basalt, we adopted the basalt characterization from Beerling et al. (2020).  
 303 The temperature was set at 22 °C. Equilibrium-based initial conditions for adsorbed and  
 304 dissolved species were established based on experimental measurements of total alkaline  
 305 cation (Ca, Mg, K, Na) quantities. The CEC was fixed at the effective CEC value (3 cmol<sub>c</sub>/kg<sub>soil</sub>)  
 306 observed at the beginning of the experiments.

307 Figure 4 presents the model-experiment comparisons for the total alkalinity release  
 308 by mineral dissolution (a), the increase in soil pH (b), and the CO<sub>2</sub> captured by the EW  
 309 applications (c). The total alkalinity release includes alkalinity observed in leaching and  
 310 soil adsorption (see Fig. S2 for the partitioning between the two phases) and constrains  
 311 the mineral weathering rates, giving  $F_D$  values in the weathering formula (10) consis-  
 312 tently below one. The pH shifts show a reasonable agreement, although the numerical  
 313 simulations do not fully reproduce the increase in pH that is observed experimentally.  
 314 This is not surprising since the model does not explain the increase in soil pH that is ob-

315 served in the experimental setup without rock application. The pH temporal dynamics  
 316 (Fig. S2) further reveal a model-experiment difference in the first days of the experiment,  
 317 where the model does not reproduce the pH experimental drop likely driven by the acid-  
 318 ity released by the cation exchange. Following the experimental work (te Pas et al., 2023),  
 319 we quantified the CO<sub>2</sub> sequestration in two ways: one based on the alkalinity liberated  
 320 through rock dissolution (potential CO<sub>2</sub> sequestration) and the other accounting for aque-  
 321 ous carbonate leaching and additional inorganic carbon stored in the soil (effective CO<sub>2</sub>  
 322 capture). Consistently with the experiments, the effective approach yielded significantly  
 323 lower estimates than the potential method because a portion of the alkalinity liberated  
 324 by the mineral dissolution is adsorbed into soil colloids, not promoting carbonate for-  
 325 mation.

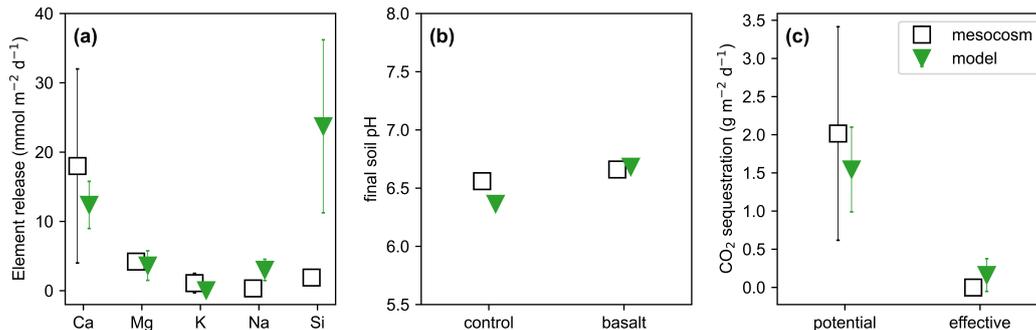
### 326 3.3 Mesocosms with Vegetation

327 The third and fourth comparisons are with mesocosm experiments (Kelland et al.,  
 328 2020; Amann et al., 2020). A distinctive feature of these experiments was the inclusion  
 329 of actively growing vegetation, specifically sorghum in Kelland et al. (2020) and wheat  
 330 and barley in Amann et al. (2020). Vegetation introduces complexities to soil hydrolog-  
 331 ogy and biogeochemistry through water transpiration, nutrient uptake, and CO<sub>2</sub> autotrophic  
 332 respiration (Appendix B). Although the representation of the hydrological cycle in these  
 333 experiments remained somewhat simplified with periodic (1-7 days) and fixed amounts  
 334 of water addition, the dynamic interplay with vegetation growth resulted in notable wa-  
 335 ter flux shifts during the growing season. This allowed for an expanded comparative anal-  
 336 ysis, including examining hydrological and soil biogeochemical processes.

#### 337 3.3.1 Reactor Columns

338 Kelland et al. (2020) conducted experiments in reactor columns measuring 16 cm  
 339 in diameter and 50 cm in depth over 120 days. We ran model simulations based on the  
 340 experimentally observed parameters as in the previous comparisons. The soil was clas-  
 341 sified as a clay loam with an initial pH of 6.6 and an organic carbon content of 1.2%. The  
 342 simulations incorporated a rainfall regime typical of temperate humid and tropical re-  
 343 gions (about 2000 mm/yr), with water added every five days. To estimate the poten-  
 344 tial evapotranspiration, we numerically recreated the experimental artificial day. This  
 345 involved maintaining photosynthetically active radiation ( $800 \mu\text{mol photons m}^{-2} \text{ s}^{-1}$ )  
 346 for 18 hr during the initial 60 days and 10 hr for the subsequent 60 days. Daily temper-  
 347 atures were computed by temporally averaging the 25 and 17 °C of the artificial day and  
 348 night, respectively. We used experimental values for the basalt application (high load  
 349 of 10 kg/m<sup>2</sup>), rock mineral composition, specific surface area, and particle size distribu-  
 350 tion. Due to the depth-averaged model framework, we could not reproduce the exper-  
 351 imental vertical heterogeneity, with basal being mixed only in the first 25 cm of the soil  
 352 column. The CEC was fixed at the experimental value of 25 cmol<sub>c</sub>/kg<sub>soil</sub>, and the ini-  
 353 tial saturation fractions were estimated based on the cation concentrations measured in  
 354 the leachate of the untreated experiment.

355 Fig. 5 presents the model-experiment comparison for the elemental release through  
 356 basalt dissolution (a), the impact on soil pH (b), and potential and effective CO<sub>2</sub> seques-  
 357 tration (c). The model well captures the release of alkaline nutrients Ca, Mg, and K. We  
 358 stress, however, that the very fast dissolution of apatite, Ca<sub>5</sub>(PO<sub>4</sub>)<sub>3</sub>(OH), comprising  
 359 around 3% of the basalt, could not be numerically reproduced (see the discussion sec-  
 360 tion) and has been added at *a posteriori* to the simulation results. The model instead  
 361 overestimates the release of Na and Si, suggesting that plants and fungi in the experi-  
 362 ments might have driven incongruent dissolution reactions. It might also be that the Si  
 363 experimental values are biased low due to underestimation of the Si pool by extraction  
 364 with ammonium acetate (Wang et al., 2004). There is also a promising model-experiment  
 365 agreement in the Ca, Mg, and Si partitioning among soil, plant, and leachate (Fig. S3).



**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 indicating  $\pm 1$  SD. (b) Final pH. (c) Potential (alkalinity release) and effective (aqueous carbonate leaching)  $\text{CO}_2$  sequestration.

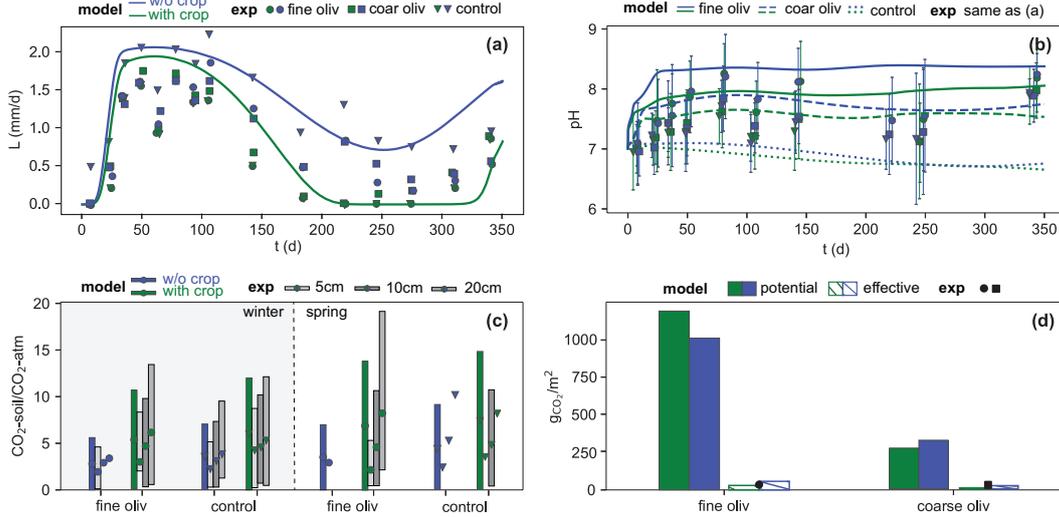
366 Moreover, the model effectively reproduces the potential  $\text{CO}_2$  sequestration resulting from  
 367 alkaline element release, demonstrating substantially higher values in both experiments  
 368 and simulations than the effective  $\text{CO}_2$  sequestered through aqueous carbonate leach-  
 369 ing.

### 370 3.3.2 Rain Barrels

371 Amann et al. (2020) performed experiments in rain barrels measuring 46 cm in di-  
 372 ameter and 26 cm in depth over a year. The soil was classified as loamy sand with an  
 373 initial pH of 7 and an initial organic carbon content of 1.2%. The simulations mimic a  
 374 rainfall regime of 800 mm/yr, with rainfall events distributed every day or week, with  
 375 little difference in the results between the two cases. The particle size distributions dif-  
 376 ferentiate between coarse and fine olivine (mostly forsterite,  $\text{Mg}_2\text{SiO}_4$ ) applications of  
 377  $22 \text{ kg/m}^2$ , with 25 and 720  $\mu\text{m}$  being the dominant diameter classes, respectively. The  
 378 simulations assume that the olivine is mixed across the barrel, while, in the experiments,  
 379 olivine was mixed in the top layer of approximately 11 cm. Temperature was varied with  
 380 a sinusoidal function across the year, from a minimum of  $6^\circ\text{C}$  to a maximum of  $25^\circ\text{C}$ .  
 381 The CEC was fixed at the experimental value of  $8.6 \text{ cmol}_c/\text{kg}_{\text{soil}}$ , initially saturated by  
 382 86.5% of Ca, 5% of Mg, 5% of K, 3% of Na, and 0.5% of H and Al (Amann et al., 2020).  
 383 We ran six simulations for the control case and the coarse and fine olivine applications,  
 384 with and without vegetation.

385 Fig. 6 presents the model-experiment comparisons in terms of hydrological balance  
 386 (a), pH dynamics (b), soil air  $\text{CO}_2$  (c), and  $\text{CO}_2$  sequestration (d). For this experimen-  
 387 tal setup, leaching is a significant proxy for hydrologic partitioning since evapotran-  
 388 spiration directly results from the difference between water input and leaching. The sim-  
 389 ulations reproduce the leaching seasonal patterns due to temperature and vegetation im-  
 390 pacts. The pH dynamics show a reasonable agreement, with an increase in the soil pH  
 391 in time due to the olivine dissolution. Both simulation and experiments show how veg-  
 392 etation tends to reduce the pH by cation uptake and  $\text{CO}_2$  respiration. The soil air  $\text{CO}_2$   
 393 dynamics also show promising results, with the model reproducing seasonal variations  
 394 and the influence of the vegetation in increasing the  $\text{CO}_2$  concentration.

395 Regarding  $\text{CO}_2$  sequestration, we estimated the potential  $\text{CO}_2$  capture due to the  
 396 Mg released by mineral dissolution and the effective  $\text{CO}_2$  capture, here quantified through  
 397 the Mg observed in the leached water, as done in the experimental work. Note that the



**Figure 6.** Model-experiment comparison based on the mesocosm experiments by Amann et al. (2020), in the presence (green) and absence (blue) of vegetation. (a) Leaching fluxes. (b) pH dynamics with error bars indicating  $\pm 1SD$ . The reported experimental values are averages between shallow (1.5 cm) and deep (24 cm) measurements. (c) Soil air  $CO_2$ , showing experimental measurements obtained at different depths and depth-averaged model results. Bars indicate  $\pm 1SD$  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.

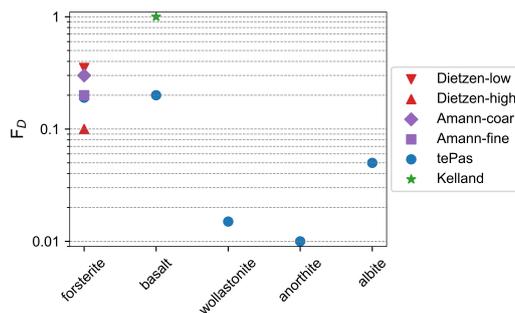
398 experimental effective  $CO_2$  capture has been reassessed from Mg leachate data due to  
 399 a dimensional inconsistency in the formula (2) reported by Amann et al. (2020). The po-  
 400 tential  $CO_2$  capture is much higher than the effective one due to the loss of alkaline cations  
 401 to soil adsorption and plant uptake. Interestingly, vegetation has a dual influence on  $CO_2$   
 402 removal: plant uptake of alkaline cations reduces the effective  $CO_2$  removal, but plant-  
 403 induced soil acidification enhances the mineral dissolution rates, increasing the poten-  
 404 tial  $CO_2$  removal. Consequently, our results suggest a trade-off in  $CO_2$  removal efficiency,  
 405 with plant-absent scenarios showcasing slower but more efficient removal processes. In  
 406 contrast, plant-present scenarios feature faster but less efficient removal processes.

## 407 4 Discussion

408 The overall favorable agreement between model outcomes and experimental observa-  
 409 tions allows us to provide critical insights into the weathering rates. We then identify  
 410 and discuss areas requiring further theoretical and experimental exploration.

### 411 4.1 Weathering Rates

412 The assessment of EW is intricately linked to the precise determination of rock weath-  
 413 ering rates, whose parameters are surrounded by considerable uncertainty (Calabrese et  
 414 al., 2022). The theoretical formulation used here (Palandri, 2004) is widely regarded as  
 415 comprehensive and is commonly applied in EW assessments (Taylor et al., 2016; Beer-  
 416 ling et al., 2020; Kantzas et al., 2022). However, this formulation is derived from exper-  
 417 imental data from stirred reactors without diffusive limitations and under conditions far  
 418 from equilibrium. In the complex, multiphase, and porous soil environment, numerous



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

419 overlooked biotic and abiotic processes may influence dissolution rates: i) concentration  
 420 gradient formation into the aqueous phase near mineral surfaces, ii) primary and sec-  
 421 ondary mineral coatings, iii) fungal and bacterial activity, iv) catalyzation or inhibition  
 422 of the dissolution reactions due to the presence of other chemical species. While some  
 423 specific processes are expected to promote dissolution (e.g., biotic activity), others pre-  
 424 dominantly impede it (e.g., particle coatings). As a result, the validity of the formula  
 425 for EW applications remains an open question.

426 As indicated by our model-experiment comparison, Palandri's formulation requires  
 427 a correction with a dissolution factor,  $F_D$  in (10), which is consistently below one for most  
 428 experimental setups and rocks (Fig. 7). In other words, minerals and rocks dissolve slower  
 429 than conventionally assumed. While these findings align with the acknowledgment that  
 430 *Actual equilibration rates are expected to be much slower than those predicted by the se-*  
 431 *lected computer code* (Palandri, 2004), they carry significant implications. Specifically,  
 432 the results underscore the need to refine weathering formulations from both a process  
 433 and data perspective, systematically incorporating other soil biotic and abiotic processes  
 434 that influence weathering. They also highlight that unless field weathering rates prove  
 435 to be much higher than what is observed in small-scale experiments, considering only  
 436 the biotic-driven enhancement of weathering rates (namely  $F_D > 1$ ) may lead to a sig-  
 437 nificant overestimation of EW  $\text{CO}_2$  removal potential (Beerling et al., 2020; Kantzas et  
 438 al., 2022).

## 4.2 Limitations and Outlook

439  
 440 The model-experiment comparison presented in this study is only a first step in the  
 441 direction of having robust assessments of EW models to reproduce observations across  
 442 scales. Many advances will be needed from both modeling and experimental perspectives.

443 From a modeling point of view, the model presented here accounts for the primary  
 444 variables of interest to assess the fate of the alkaline cations released by the EW appli-  
 445 cations and their corresponding inorganic carbon sequestration potential. It does not in-  
 446 clude feedback that the rock powder application may have on some soil physical and bi-  
 447 otic processes. The different leaching fluxes observed in the experiments by (Amann et  
 448 al., 2020) suggest that the rock powder impacts the soil texture and hydraulic conduc-  
 449 tivity, hence the soil water partitioning. Specific experiments evaluating the temporal  
 450 evolution of soil physical properties are needed to incorporate such feedback in models,  
 451 even though some theoretical estimates may be derived based on soil physics models (Jury  
 452 & Horton, 2004). Furthermore, rock applications may influence biotic activity and the  
 453 organic carbon balance, with potentially detrimental effects in tropical soils and peat-

454 lands (Klemme et al., 2022). Modeling advances could also include mass balances for heavy  
455 metal accumulations, such as nickel and copper, which are significant concerns in the con-  
456 text of EW applications (Haque et al., 2020; Dupla et al., 2023)

457 Compared to the more used reactive transport models, our model is simpler, not  
458 accounting for soil vertical heterogeneity and including fewer chemical species. This sim-  
459 plicity comes at the cost of spatial information but at the advantage of accessibility and  
460 a more concentrated examination of temporal dynamics. Interestingly, our model did not  
461 necessitate a semi-empirical pH buffer function commonly employed in EW simulations  
462 with reactive transport models to avoid unrealistic spikes in soil pH. This is intriguing  
463 since our buffering mechanisms incorporate only carbonate chemistry and cation adsorp-  
464 tion while neglecting others like organic alkalinity. We attribute this success to imple-  
465 menting shorter simulation time steps, operating in the order of minutes rather than days,  
466 as seen in traditional reactive transport models. Nonetheless, our model encounters lim-  
467 itations in reproducing the rapid dissolution of certain minerals and materials (e.g.,  $\text{Ca}_5(\text{PO}_4)_3(\text{OH})$   
468 or  $\text{Ca}(\text{OH})_2$ ) due to pronounced spikes in alkalinity that hinder the numerical conver-  
469 gence of the implicit system (Appendix A). Looking ahead, a promising avenue involves  
470 integrating our model results with both reactive transport models and observational data  
471 to gain comprehensive insights into soil EW dynamics.

472 From an observational standpoint, it is worth acknowledging the temporal constraint  
473 within the available datasets. Specifically, only one of the experimental datasets used  
474 within this study spans a complete year (Amann et al., 2020), while others have a rel-  
475 atively shorter duration of a few months. Given the potential yearly timescales associ-  
476 ated with the dissolution of EW rock powder, extrapolating results becomes challeng-  
477 ing, especially considering that weathering rates may decrease over the years (Calabrese  
478 et al., 2022). Additionally, most experiments relied on elevated rock loadings (i.e.,  $\geq 10$   
479  $\text{kg/m}^2$ ) to enhance signals within the short experimental time frame, although such load-  
480 ings may not be realistic for practical applications. There is also an opportunity to ex-  
481 plore the influence of realistic stochastic rainfall regimes, which is often absent in cur-  
482 rent experimental setups. Integrating field observations of large-scale EW applications  
483 will hopefully address some of these temporal and loading limitations, offering insights  
484 into the compatibility between model results, small-scale experiments, and the practi-  
485 cal considerations of large-scale field trials.

## 486 5 Conclusions

487 While enhanced weathering (EW) holds great promise as a negative emission strat-  
488 egy, thanks to its significant CDR potential, low technological prerequisites, and valu-  
489 able co-benefits, no model has been shown to reproduce EW observations at scale. This  
490 deficit restricts our ability to make accurate quantitative predictions for assessments of  
491 CDR via EW. In this study, we took a benchmark step in this direction, advancing a rel-  
492 atively accessible ecohydrological and biogeochemical model whose results could be metic-  
493 ulously compared with four distinct experimental datasets of different complexity.

494 The model-experiment comparison demonstrates an overall favorable agreement  
495 for the primary variables of interest, including alkalinity release, pH dynamics, and  $\text{CO}_2$   
496 sequestration. The comparison also demonstrates that weathering rates are lower than  
497 traditionally assumed by one or two orders of magnitudes and highlights further research  
498 directions to improve our understanding and quantitative predictive power for EW as  
499 a NET. Finally, while representing EW dynamics within the soil’s upper layers is cru-  
500 cial, EW negative emission potential is linked to the fate of rock dissolution products  
501 from the field to the ocean, a journey yet to be fully disclosed.

502 **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 control volume are distributed among the different soil phases. Specifically, alkaline cations ( $X_{\text{tot}}$ ) are distributed between dissolved and adsorbed phases, inorganic carbon ( $IC_{\text{tot}}$ ) is distributed between aqueous and air phases, and aluminum ( $Al_{\text{tot}}$ ) exists dissolved in water, adsorbed to the soil matrix or in complexes with organic or inorganic matter. In formula

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

$$IC_{\text{tot}} = n_Z s[\text{DIC}] + n_Z r(1-s)[\text{CO}_2]_a, \quad (\text{A2})$$

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

503 where  $n_X$  is the cation valence and CEC is the cation exchange capacity. The latter in-  
504 dicates the moles of dissolved cations that can be adsorbed on soil colloids due to their  
505 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  $\text{Ca}^{2+}$  with  $\text{Al}^{3+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{H}^+$ :

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

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

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

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

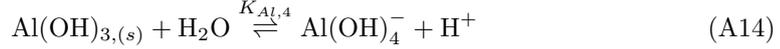
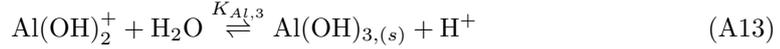
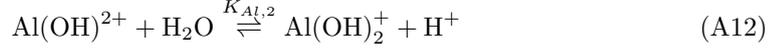
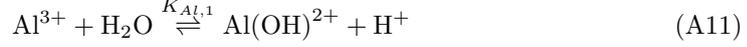
$$[\text{HCO}_3^-] = K_1 [\text{CO}_2]_w / [\text{H}^+], \quad [\text{CO}_3^{2-}] = K_1 K_2 [\text{CO}_2]_w / [\text{H}^+]^2, \quad (\text{A9})$$

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

506 where  $K_1$  and  $K_2$  are the first and second carbonic acid dissociation constants.  $K_w$  is  
507 the water dissociation constant.  $K_{\text{H}}$  is Henry's constant for  $\text{CO}_2$  solubility. All these con-  
508 stants and their temperature dependence are evaluated after (Stumm & Morgan, 1996).  
509 The combination of Eqs. (A4) and (A7) summarize EW goal of increasing alkalinity by  
510 mineral dissolution to promote aqueous carbonate formation. The efficiency of this pro-  
511 cess varies as a function of the water chemistry (Bertagni & Porporato, 2022). Formal

512 extension to the alkalinity definition (A7) could include aluminum, which plays a buffer  
 513 role in acidic conditions, and organic alkalinity. Other weak acids and bases have been  
 514 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 systems 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  $\text{Al}^{3+}$  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

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

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

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

## 516 Appendix B Plants dynamics and their role in EW

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). In the model, growing vegetation ( $V$ ) can be dynamically modeled through a classical logistic equation

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

517 where  $k_V$  is the carrying capacity per unit area, dependent on plant and ecosystem types,  
 518 and  $\alpha_V$  is the plant growth rate. The growth rate can be estimated based on the time  
 519 ( $t_V$ ) required for plants to progress from seedling to maturity through  $\alpha_V \approx 6/t_V$ . Sim-  
 520 ulated plant-mediated processes are then scaled with the normalized vegetation variable  
 521  $\hat{V} = V/k_V$ , defined between 0 and 1. These processes include: i) plant transpiration  
 522 ( $T$ ), modeled as a soil moisture function (Laio et al., 2001a); ii) plant passive uptake,  
 523 assumed to be directly proportional to the transpiration rate (Cipolla et al., 2021a); iii)  
 524 autotrophic respiration ( $\text{RESP}_a$ ), estimated to be equivalent to heterotrophic respira-  
 525 tion when plants are fully grown (Bond-Lamberty et al., 2004); iv) active uptake, which  
 526 occurs during plant growth when passive uptake alone cannot meet the nutrient demands  
 527 for growth (see below). Noteworthy, both plant-mediated nutrient uptake and inorganic  
 528 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; 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  $\ell$  is the typical distance traveled from bulk to root. The latter can be quantified as  $\ell = \sqrt{r_d Z / (\hat{V} \cdot \text{RAI})}$ , 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

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

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

533 **Data and Software Statement** The numerical codes for the EW model (Python),  
534 the Jupyter Notebooks for the model-experiment comparisons, and all numerical data  
535 produced within this manuscript are available at [https://github.com/MatteoBertagni/](https://github.com/MatteoBertagni/EWmodel)  
536 `EWmodel`. The experimental data come from previous publications, as acknowledged in  
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