

1 **Enhanced weathering strategies for stabilizing climate and averting**
2 **ocean acidification**

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15 Chemical breakdown of rocks, ‘weathering’, is an important but very slow part of the
16 carbon cycle that ultimately leads to CO₂ being locked-up in carbonates on the ocean
17 floor. Artificial acceleration of this carbon sink via distribution of pulverized silicate
18 rocks across terrestrial landscapes may help offset anthropogenic CO₂ emissions¹⁻⁵. We
19 show that idealized enhanced weathering scenarios over less than a third of tropical land
20 could significantly drawdown atmospheric CO₂ and ameliorate ocean acidification by
21 2100. Global carbon cycle modelling⁶⁻⁸ driven by ensemble Representative
22 Concentration Pathway (RCP) projections of 21st century climate change (RCP8.5,
23 business-as-usual; RCP4.5, medium-level mitigation)^{9,10}, indicates that enhanced
24 weathering could lower atmospheric CO₂ by 30-300 ppm by 2100 depending mainly on
25 silicate rock application rate (1 kg or 5 kg m⁻² yr⁻¹) and composition. At the higher
26 application rate, end-of-century ocean acidification is reversed under RCP4.5 and
27 reduced by about two-thirds under RCP8.5. Additionally, surface ocean aragonite
28 saturation state, a key control on coral calcification rates, is maintained above 3.5
29 throughout the low latitudes, thereby helping maintain the viability of tropical coral reef
30 ecosystems¹¹⁻¹⁴. However, we highlight major issues of cost, social acceptability, and
31 potential unanticipated consequences that will limit utilization and emphasize the need
32 for urgent efforts to phase down fossil fuel emissions¹⁵.

33 In 1992, over 170 nations agreed to limit anthropogenic CO₂ emissions to avoid ‘dangerous’
34 human-made climate change¹⁶, yet massive expansion of fossil fuel extractions, including
35 shale gas and tar sands, is allowing emissions to grow¹⁷. Avoiding dangerous climate change
36 may therefore require the controversial deployment of Carbon Dioxide Removal (CDR)
37 schemes^{4,18}, so called ‘negative emissions’ strategies whereby CO₂ is captured and removed
38 from the atmosphere. The Fifth Assessment Report of the Intergovernmental Panel on
39 Climate Change¹² and the U.S. National Research Council Report¹⁸ both recognized enhanced
40 terrestrial weathering of silicate rocks as an important but poorly constrained CDR approach.
41 Currently, natural weathering of silicate and carbonate rocks consumes ~0.25 Pg C yr⁻¹ of
42 atmospheric CO₂, which is ~3% of fossil fuel emissions¹⁹ (~9–10 Pg C yr⁻¹). Artificially
43 accelerating this land-based CO₂ sink involves the intentional application of pulverised
44 silicate rocks to vegetated landscapes to markedly enhance CO₂ consumption¹⁻⁵. However,
45 assessments to date have excluded primary drivers of soil mineral weathering, especially
46 terrestrial ecosystem processes and feedbacks from CO₂ and future climate change, limiting
47 our understanding of its capacity to offset fossil fuel CO₂ emissions¹².

48 Here we present spatially resolved analyses of enhanced weathering by terrestrial
49 ecosystems as a macro-engineering CDR option based on idealized cases for distributing
50 pulverised silicate rocks in the tropics using multi-model ensemble projections (Coupled
51 Model Intercomparison Project, CMIP5) of 21st century climate change^{9,10}. Our modelling
52 framework includes climate-plant-soil linkages important for regulating mineral weathering
53 by coupling a detailed weathering model with a dynamic global vegetation model and
54 accounting for land surface hydrology, topography and lithology^{6,7} (Methods). We assess
55 effects of enhanced weathering on net CO₂ consumption and examine feedbacks on
56 atmospheric CO₂ and ocean chemistry over the next century using a suite of five CMIP5
57 general circulation model (GCM) simulations (1°lat. × 1°lon.)^{9,10} for each of two
58 Representative Concentration Pathway scenarios (RCPs): RCP8.5 (business-as-usual), and
59 RCP4.5 (medium-level stabilization of emissions); postscripts (8.5 and 4.5) denote radiative
60 forcing (W m⁻²) in year 2100 relative to year 1750 (Supplementary Information).
61 Assessments are undertaken for various application rates of the igneous rocks dunite (>90%
62 olivine, Mg₂SiO₄) (Ref. 3) and harzburgite (50–90% olivine), which are both commercially
63 mined, and basalt for which major resources exist in terrestrial large igneous provinces
64 (LIPs)²⁰ (Fig. 1). These rates fall within the range adopted in the early 1930s for rejuvenating
65 European forest soils with basalt to encourage tree growth²¹.

66 Our simulations indicate that terrestrial weathering can be markedly increased by
67 distributing pulverised silicate rocks throughout the tropics (30°N to 30°S), potentially
68 consuming hundreds of petagrams (1×10^{15} g) of CO₂ by 2100 (Fig. 1). Ensemble median
69 CO₂ consumption by terrestrial weathering increases towards a maximum as the total rock
70 applied increases, with olivine-rich dunite and harzburgite being about twice as effective as
71 basalt for equivalent application rates (Fig. 1a–c). We present CO₂ consumption curves
72 assuming mixing depths of 10 cm and 30 cm for each application rate; 10 cm is likely the
73 minimum mixing depth given intense precipitation events, the distribution of macropores and
74 bioturbation by invertebrates in tropical soils down to depths of 30–50 cm (Supplementary
75 Information). In the model, CO₂ consumption by weathering increases when added rock
76 grains mix deeper in the soil, particularly at the $5 \text{ kg m}^{-2} \text{ yr}^{-1}$ application rate, because mineral
77 saturation, a chemical brake on weathering, occurs more slowly in a larger soil solution
78 volume. Overall CO₂ consumption patterns for a particular RCP scenario show a consistently
79 narrow range of variation across the five ensemble GCMs (Fig. 1d–f). For a given application
80 rate, the magnitude of CO₂ consumption is similar for the business-as-usual (RCP8.5) and
81 medium level mitigation (RCP4.5) scenarios (Fig. 1d–f), largely because the runoff for the two
82 scenarios is similar (Supplementary Information).

83 Comparing cumulative end-of-century amounts of pulverised rock added to the tropics
84 with estimated total resources indicates dunite has limited utility for long-term atmospheric
85 CO₂ removal³ (Fig. 1), whereas sufficient harzburgite and basalt resources exist for the
86 application rates considered here (Fig. 1, Supplementary Information). The rock mass
87 required can be reduced by restricting application to regional intense tropical weathering
88 ‘hotspots’ (Fig. 1, Supplementary Information). Such optimization reduces the land area
89 required by more than two-thirds, from 69 Mkm² to 20 Mkm², and total rock mass by 70%,
90 whilst still achieving ~80–89% of the effect (Fig. 1a–c, symbols). Hotspot land areas are
91 primarily tropical forests except parts of Asia which are croplands. However, basalt can
92 promote crop growth on highly weathered acidic tropical soils^{22,23} by increasing soil
93 alkalinity, cation exchange capacity and the availability of growth-limiting phosphorus, with
94 associated reductions in Al and Mn toxicity^{23,24}. Ample basalt resources exist within the
95 major LIPs in the tropics (Ethiopian Traps, Deccan Traps and Paraná Traps) to support
96 simulated application rates (Fig. 1) and these sources could exploit existing infrastructure for
97 distribution. Meeting silicate rock demand would require large-scale mining operations, e.g.,

98 throughout the major tropical LIPs, with production rates exceeding those for coal and
99 adverse consequences for local ecosystems.

100 As CO₂ is removed from the atmosphere by enhancement of the weathering carbon sink,
101 the carbon cycle responds by redistributing carbon among surface reservoirs (atmosphere,
102 ocean, soil, and land biosphere), with CO₂ out-gassing by the ocean in particular offsetting
103 some of the artificial drawdown¹⁷. There is, consequently, a ‘rebound’ effect whereby each
104 extra mole of CO₂ consumed does not translate into the removal of a mole of atmospheric
105 CO₂ over time. We therefore estimate the effects of our CO₂ consumption fluxes on the
106 RCP4.5 and RCP8.5 atmospheric CO₂ trajectories through the 21st century with the well-
107 tested GENIE Earth system model⁸ that broadly captures these responses. Distributing 1kg m⁻²
108 yr⁻¹ of pulverised silicates across 20 Mkm² of tropical weathering ‘hotspots’ lowers
109 atmospheric CO₂ concentrations by ~40 ppm (basalt) or ~140 ppm (harzburgite) by year 2100
110 in both the RCP4.5 and RCP8.5 climate change scenarios (Fig. 2a, b). Increasing the
111 application rate to 5 kg m⁻² yr⁻¹ over the same 20 Mkm² ‘hotspot’ areas lowers the
112 atmospheric CO₂ concentration further by 150–180 ppm under both RCPs (Fig. 2c, d), with
113 an increasing effect at deeper soil mixing depths. For RCP4.5, atmospheric CO₂ by 2100 is
114 reduced from 540 ppm to 390–350 ppm (basalt) or 350–250ppm (harzburgite), sufficient to
115 play a major role in stabilizing climate and avoid seeding long-term amplifying climate
116 feedbacks¹⁷ (Fig. 2). For the business-as-usual RCP8.5 scenario, however, the lowest
117 simulated CO₂ concentration by year 2100 in the high-end weathering scenario is still ~730
118 ppm (basalt) or 690–560 ppm (harzburgite) (Fig. 2d). This suggests even massive
119 intervention in Earth’s carbon cycle with basalt is unable to drive atmospheric CO₂ down
120 close to the target of 350 ppm by 2100, an estimated requirement for restoring planetary
121 energy balance and stabilizing climate¹⁷.

122 Future climate warming averted (WA) by engineering CO₂ removal through enhanced
123 weathering is dependent on climate sensitivity and the actual atmospheric CO₂ concentration.
124 Calculated end-of-century ‘warming averted’ figures for the enhanced weathering scenarios
125 using GENIE, which has a low-to-medium climate sensitivity, are summarized in Table 1.
126 For high application rates, WA ranges from 0.9–2.2°C for RCP4.5 and 0.7–1.6°C for RCP8.5
127 (Table 1). At low application rates, corresponding ranges of WA are 0.2–0.7°C for both
128 RCPs (Table 1). These numbers suggest that, theoretically at least, negative emissions from
129 enhanced weathering could play a role alongside conventional mitigation reducing net CO₂
130 emissions in limiting future warming²⁵.

131 Unmitigated future increases in atmospheric CO₂ will not only drive climate change but
132 also ocean acidification, including reduced saturation of surface waters with respect to
133 aragonite, threatening reef-building coral ecosystems¹¹⁻¹⁴. Artificially enhanced tropical
134 weathering increases land-to-ocean fluxes of alkalinity and dissolved inorganic carbon and
135 raises freshwater pH to the upper range of tropical rivers (Supplementary Information). These
136 fluxes, together with reduced atmospheric CO₂ (Fig. 2), tend to counter the negative impacts
137 on ocean carbonate chemistry (Figs. 3 and 4). Our simulations driven by decreased CO₂ (Fig.
138 2) and increased alkalinity fluxes show that additions of 1 kg m⁻² yr⁻¹ of harzburgite or basalt
139 across the weathering ‘hotspots’ can mitigate future ocean acidification by an average of
140 around 0.1 pH units (Fig. 3a, b). A higher silicate application rate (5 kg m⁻² yr⁻¹) reverses
141 future surface ocean acidification under RCP4.5, restoring global mean surface ocean pH to
142 year 2000 levels or even pre-industrial levels by 2100 (Fig. 3c). Even for RCP8.5, 5 kg m⁻²
143 yr⁻¹ reduces ocean acidification by approximately two-thirds by year 2100 (Fig. 3d)
144 (Supplementary Information).

145 Coral reef health is linked to the ocean’s aragonite saturation state (Ω_a), which affects the
146 rate at which corals can precipitate this crystalline mineral form of calcium carbonate and
147 build skeletons^{13,14}. Modern coral reefs generally occur where open ocean waters have a
148 value of Ω_a above a postulated¹⁴ critical threshold of ~3.5. But under RCP4.5, and especially
149 RCP8.5, Ω_a at reef sites drops to <3.5 by 2100 (Fig. 4), potentially threatening them with
150 extinction¹⁴. In simulations for RCP4.5 and RCP8.5, enhanced weathering with 1 kg m⁻² yr⁻¹
151 of silicates (basalt or harzburgite) and reduced atmospheric CO₂, generates conditions of Ω_a
152 >3.5 across main regions of coral reef occurrence (Fig. 4a–e). Hence, although this low
153 dosage is rather ineffective at reducing global CO₂ (Fig. 2), it has specific regional advantages
154 in terms of helping protect coral reefs. Applications of either rock at high rates (5 kg m⁻² yr⁻¹)
155 markedly increase Ω_a above 3.5 in both RCP4.5 and RCP8.5 scenarios at low latitudes (Fig.
156 4c,f). Enhanced weathering on land could therefore be more effective at alleviating stressors
157 on coral reef health, including ocean acidification, than enhanced open-ocean dissolution of
158 olivine^{26,27}.

159 Our spatial and temporal analyses incorporate detailed plant-soil-climate interactions
160 regulating soil mineral weathering rates. Driven by detailed geographical variations in
161 projections of 21st century climate change and vegetation activity, they indicate the maximum
162 potential of enhanced weathering for climate change mitigation, including amelioration of

163 ocean acidification. However, our scenarios represent a suite of idealized cases in which
164 application of pulverised silicate rocks over forests is assumed to be achievable over large
165 regions. Consequently, they help define the maximum potential CDR capacity of the
166 approach. Not only will practical barriers to mineral transport and distribution on biodiverse
167 tropical forests limit large-scale deployment, but roll-out on such a large-scale may be
168 undesirable from both conservation and ecosystem services viewpoints. Deployment might
169 be achievable in areas undergoing reforestation/afforestation or on agricultural lands where
170 existing infrastructure could be utilized for rock grain distribution and management.
171 However, well-documented field studies on graded spatial scales are needed prior to any
172 significant implementation.

173 Large-scale geoengineering is ethically fraught¹⁵ and poses dangers of both foreseeable
174 and unforeseen consequences. Enhanced weathering employs naturally occurring minerals
175 and reactions and therefore falls in the category of “soft geoengineering” along with
176 reforestation, and agricultural techniques increasing soil carbon storage²⁸. Nevertheless, it
177 still requires comprehensive environmental impact assessments and dust mitigation strategies
178 at production and deployment sites. Additionally, the production and distribution of
179 pulverised rock carries health risks to anyone coming in contact with it because the particle
180 sizes involved are respirable (Supplementary Information). Harzburgite, for example,
181 includes asbestos-related minerals that carry health risks to local populations near application
182 sites. However, carefully implemented, enhanced weathering may have added benefits,
183 including fertilizing ocean and terrestrial CO₂ capture by marine diatoms^{3,26,29} and tropical
184 forests, respectively. Such effects, which are not considered here, could help offset energy
185 costs^{3,5} associated with extensive rock mining, grinding and transportation operations that
186 might lower its sequestration capacity by ~8–33%.

187 Estimated implementation costs (combined capital and operational) for achieving an
188 initial 50 ppm drawdown of atmospheric CO₂ are \$60–600 trillion for mining, grinding and
189 transportation, assuming no technological innovation, with similar associated additional costs
190 for distribution (Supplementary Information). On this basis, costs of enhanced weathering as
191 a ‘negative emissions’ option exceed an estimate of \$50–200 trillion¹⁷ for air capture of 50
192 ppm CO₂, but with the latter being less effective in reducing ocean acidification in important
193 coral reef regions. These issues support calls for the alternative of a rising international
194 carbon fee¹⁷. We proffer enhanced weathering not as a panacea for erasing impacts of fossil

195 fuel burning, but as a sobering indication of actions that may be required if fossil fuel
196 emissions are not phased-down rapidly.

197 **Methods**

198 Methods and any associated references are available in the online version of the paper.

199

200 **References**

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291 and A.R. provided model set-up support and advice, M.R.L. analysed the CMIP5 climates. D.J.B. led
292 the writing with contributions from all co-authors, especially J.H., A.R., J.Q. and L.L.T.

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Figure Legends

297 **Figure 1. Enhanced weathering from pulverised silicate rock additions to the tropics**
298 **increases CO₂ consumption.** End-of-century CO₂ consumption by enhanced terrestrial
299 weathering with (a) dunite, (b) harzburgite or (c) basalt as a function of total rock applied,
300 defined as the product of rate and an increasing treated land area in the tropics (30°S to 30°N).
301 Simulations are shown for the Representative Concentration Pathway (RCP) 8.5; median and
302 range for five climate model simulations for each application scenario. Symbols indicate
303 reductions in CO₂ consumption and total rock applied when application is limited to 20 Mkm²
304 of tropical weathering hotspots; symbol shape and fill denotes application rate and mixing
305 depth scenario, respectively, for each curve. Vertical red lines show estimated total resources
306 for each rock type (for basalt, solid, dashed and dot-dashed lines represent basalt resources in
307 each of the Ethiopian, Deccan and Paraná Traps, respectively). The shaded area denotes
308 uncertainty in upper values of global dunite resource availability (Supplementary
309 Information). Panels (d) to (f) display the corresponding ensemble ranges for high and low
310 application scenarios for each of five climate model simulations for both RCP8.5 and RCP4.5,
311 assuming a mixing depth of 30 cm.

312 **Figure 2. Enhanced weathering lowers atmospheric CO₂ with projected 21st century**
313 **climate change.** Effects of low additions (1 kg m⁻² yr⁻¹) of silicate rock to 20 Mkm² of
314 tropical weathering hotspots for two mixing depths (10 cm and 30 cm) on the atmospheric
315 CO₂ concentration for (a) RCP4.5 (medium-level mitigation) and (b) RCP8.5 (business-as-
316 usual). Panels (c) and (d) show comparable results for the effect of higher additions (5 kg m⁻²
317 yr⁻¹) of silicate rocks to the same areas of the tropics. In all panels, the pale blue line indicates
318 the ‘control’ run with RCP-driven weathering, without additions of silicate rock. Envelopes
319 and lines (solid 10 cm/dashed 30 cm) show the smoothed (five-year boxcar) ranges and
320 medians, respectively, of results from five climate models for each RCP.

321 **Figure 3. Enhanced weathering ameliorates future ocean acidification caused by**
322 **projected 21st century increases in atmospheric CO₂.** Effects of increased alkalinity fluxes
323 resulting from additions of 1 kg m⁻² yr⁻¹ of silicate rock to 20 Mkm² of tropical weathering
324 hotspots mixed to two soil depths on global surface ocean pH for (a) RCP4.5 (medium-level
325 mitigation) and (b) RCP8.5 (business-as-usual). Panels (c) and (d) show comparable results
326 for the effect of higher additions (5 kg m⁻² yr⁻¹) of silicate rocks to the tropics on global
327 surface ocean pH for RCP4.5 and RCP8.5 respectively. Envelopes and lines (solid 10
328 cm/dotted 30 cm mixing depths) show the smoothed (five-year boxcar) ranges and medians,
329 respectively, of results from five climate models for each RCP.

330 **Figure 4. Enhanced weathering raises the aragonite saturation state of the ocean by**
331 **2100.** Simulated global distribution of the aragonite saturation state (Ω_a) of the surface ocean
332 in 2100 for RCP4.5 (a) no addition of silicate rocks, 1 kg m⁻² yr⁻¹ of (b) basalt and (c)
333 harzburgite distributed over the tropics. Corresponding simulations for RCP8.5 are given in
334 (d), (e) and (f) for applications of 5 kg m⁻² yr⁻¹. Each panel displays in black the distribution
335 of reef-building corals (www.reefbase.org). All simulations are for a mixing depth of 30 cm.

336 **Methods**

337 **Terrestrial rock weathering modelling.** Terrestrial vegetation delivers the carbon-energy
338 flux in the form of photosynthate to roots and associated mycorrhizal fungal networks which
339 fuels biotic rates of mineral dissolution³⁰. In our rock weathering model, the sub-surface
340 organic carbon flux is stoichiometrically coupled to the rate of primary production and the
341 uptake of inorganic nutrient ions by roots and mycorrhizal fungi which regulate the ionic
342 composition and charge balance of the microscopic region of soil pore fluids at the organism-
343 mineral interface (the mycorrhizosphere)^{6,7}. This balance controls local pore fluid pH and
344 organic ligand concentrations at the reacting mineral surfaces that control the rates of mineral
345 dissolution through well-described reaction mechanisms³¹. We therefore couple an extended
346 version of a previously published rock weathering model^{6,7} with the Sheffield Dynamic
347 Global Vegetation Model (SDGVM)³². Our simulations employed fixed land use patterns³³.
348 The SDGVM simulates terrestrial carbon, nitrogen and water cycling by vegetation and soils
349 including land surface net primary productivity (NPP), hydrology, autotrophic and
350 heterotrophic soil respiration, and dissolved organic carbon pools³². SDGVM is comparable
351 in its sensitivity of response to CO₂ and climate to other DGVMs^{34,35}.

352 In the extended weathering model, rainwater with an initial pH determined by the
353 partial pressure and solubility of atmospheric CO_{2(g)} and ion charge balance percolates
354 through the soil at a rate determined by the SDGVM runoff. Soil solution chemistry is
355 calculated both within and outside the mycorrhizosphere within the soil profile, which is
356 divided into 10 layers specified at increasing depths, with runoff composition from each layer
357 mixed and advected into the next layer. Mixing of bulk soil and mycorrhizosphere water is
358 conceptualised as, but not explicitly parameterised as, hydrodynamic dispersion and diffusive
359 exchange of bulk soil fluid solutes with the decreasing mycorrhizosphere volume with depth⁶,
360 and pore fluid transport to plant roots for transpiration. The model recalculates the soil
361 solution chemistry of each layer, with the dissolution reaction progress of primary silicate
362 minerals ceasing upon reaching the theoretical saturation state of the fluid with respect to the
363 dissolving mineral. Thermodynamic equilibria constrain both the forward reaction for
364 mineral dissolution (see Eq. 1 below) and the concurrent precipitation of secondary phases
365 including kaolinite, gibbsite and amorphous silica which act as sinks for dissolved Al and Si
366 released by weathering. On carbonate-bearing lithologies, pore fluids are equilibrated with
367 any calcite, dolomite or gypsum which might be present before weathering of any silicate

368 minerals present takes place. This treatment assumes sufficient carbonates to maintain
 369 solubility equilibrium during the simulation time horizon and is not suitable for trace amounts
 370 of carbonate minerals which would become completely depleted in non-carbonate lithologies.
 371 Soil solution chemistry, therefore, depends on solubility equilibrium with existing carbonate
 372 or sulphate minerals, precipitation of secondary phases, including kaolinite and amorphous
 373 silica, and weathering of primary silicate minerals.

374 Underlying the weathering model is a rasterised version of the Hartmann & Moosdorf³⁶
 375 lithological map for which we prepared a lithological database giving the proportions of the
 376 parent minerals in each rock type. Each rock type has its own mineral assemblage (see
 377 below), and each mineral m weathers according to the general rate law³⁷ with mineral-
 378 specific parameter values for SA , k , n and E :

$$379 \quad Rate_m = SA_m \sum_i \left[k_{i,m}^{298.15} \exp \left[\frac{-E_{i,m}}{R} \left(\frac{1}{T} - \frac{1}{298.15} \right) \right] a_i^{n_{i,m}} \left(1 - \left[\frac{Q_m}{K_{sp,m}} \right] \right) \right] \quad \text{Eq. (1)}$$

380 where $Rate_m$ is given in mol m⁻² mineral s⁻¹, SA is mineral surface area (m²), i is the individual
 381 weathering agent, such as [H⁺], $k_{i,m}$ is the rate constant, $E_{i,m}$ is the apparent activation energy
 382 (kJ mol⁻¹), R is the gas constant (kJ mol⁻¹ K⁻¹), T is temperature (K), a_i is the molar activity of
 383 weathering agent i (mol l⁻¹) and $n_{i,m}$ is the reaction order. $Q_m = \prod_j a_j^{s_j}$ is the ion activity
 384 product of the soil solution, where a_j is the activity of solute j raised to the power of its
 385 stoichiometry s_j on the product side of the chemical equation describing the dissolution of
 386 mineral m (Tables S2, S3). $K_{sp,m}$ is the solubility constant for mineral m (Table S2).
 387 Activities are approximated by concentrations.

388 The model accounts for changes in mineral surface area due to relief (standard deviation
 389 of orography) as described previously⁷ and includes an empirical surface area correction for
 390 each rock type which accounts for age effects³⁸, internal porosity, grain size errors and
 391 deviation of particle shape from perfect spheres. Soil water residence times and riverine
 392 fluxes depend on run-off calculated by SDGVM. The model calculates monthly fluxes of
 393 CO₂ consumption, alkalinity (Ca²⁺, Mg²⁺, K⁺ and Na⁺) and dissolved inorganic carbon and is
 394 verified against water chemistry and discharge data from a global suite of river catchments

395 **Validation.** Simulated terrestrial fluxes of CO₂ consumed by rock weathering in 42
 396 watersheds worldwide³⁹ using the CRU-3 climate⁴⁰ at 1° × 1° resolution are validated against
 397 fluxes derived from catchment-scale estimates based on stream-water chemistry

398 (Supplementary Information). We generated fluxes for basins in the World Resources
399 Institute's shapefile⁴¹ and compared these to the 45 basins (Table 3 in Ref. 39) where the
400 basin names could be matched. Given the test aims to compare model and observed
401 weathering rates, three catchments were rejected on the basis of markedly different basin
402 areas or runoff (defined as two standard deviations of the mean residual). Therefore, 42
403 basins were retained for validation, except in the case of carbonate CO₂ consumption, where
404 an additional five basins were excluded due to lack of carbonates in the modelled lithologies.

405 The response of modelled weathering rates in Iceland to temperature change accords with
406 observed chemical weathering flux responses to climate warming over the past four decades
407 in un-glaciated Icelandic catchments⁴². Regional responses also support the CO₂ and climate
408 change sensitivity of our approach. Adopting a more complex soil weathering module
409 coupled to a DGVM⁴³, another group predicted a similar increase in CO₂ consumption in the
410 Mackenzie River arctic watershed from 355 ppm CO₂ and modern climate to 560 ppm CO₂
411 with an associated warmer climate, based on results from the RCP4.5 simulations
412 (Supplementary Information). Estimated pH values of river run-off calculated from alkalinity
413 fluxes and equilibrated to ambient CO₂, are comparable to measurements reported for a range
414 of tropical river catchments (Supplementary Information).

415 **Geoengineering simulations.** For the atmospheric CO₂ concentration trajectories defined by
416 the two RCPs considered here (RCP4.5 and RCP8.5), the five General Circulation Models
417 (GCMs) produce monthly temperature, relative humidity and precipitation to drive SDGVM.
418 Monthly climate datapoints closest to the desired coordinates are bilinearly interpolated in
419 space before daily values for the month are estimated using climate statistics. These
420 estimated daily climates force SDGVM. Distributed silicate rock grains are treated as perfect
421 monomineralogical spheres with a nominal starting diameter of 10 μm. Initial total surface
422 areas for the added silicates are calculated for each mineral using the total mass applied,
423 specified diameter, weight fraction for the mineral, mineral specific gravity and the equations
424 for the volume and surface area of a sphere. As weathering progresses, mass is removed and
425 the fractional change in total surface area is estimated using the fractional change in mass
426 raised to the power $\frac{2}{3}$. This treatment assumes that each particle is a shrinking sphere. The
427 total mass and surface area for each mineral are increased according to dose rate. No attempt
428 is made to model a particle size distribution for either the starting silicate rock grain size or
429 the individual mineral residues following weathering. Pulverized silicates are mixed with a

430 specified depth of soil, without modelling bioturbation processes or the transport behaviour of
431 suspended materials in infiltrating water. Soil water residence times and riverine fluxes
432 depend on runoff modelled by SDGVM. Mean reactive surface areas of autochthonous
433 primary soil minerals are corrected for erosion and relief⁷. The model assumes no change in
434 porosity or water movement with depth, and there is no preferential transport of different
435 particle sizes.

436 **Mineralogy of pulverized silicates.** The mineralogy of each simulated pulverized silicate
437 rock is listed in Table S4. The model basalt silicate mineralogy is based on the normative
438 composition for a normal alkali tholeiitic basalt⁴⁴, neglecting some minor phases such as
439 magnetite. Our dunite composition follows Kogel *et al.*⁴⁵. We use the mineralogy of the
440 Troodos harzburgite, with lizardite rather than chrysotile (asbestos) as this is the dominant
441 serpentine near Troodos⁴⁶ and as it is sensible to avoid rocks with a large proportion of
442 asbestos for health reasons. Our results are therefore conservative with respect to the
443 proportion of unserpentinised olivine and the relative amounts of lizardite and chrysotile
444 present.

445 **GENIE Earth system global CO₂ and ocean biogeochemistry modelling.** The climate and
446 ocean circulation of the GENIE Earth system model has been calibrated by 2-D reanalysis
447 fields of surface air temperature and humidity and 3-D observational fields of ocean
448 distributions of temperature and salinity⁴⁷. The carbon cycle is calibrated against observed
449 ocean phosphate and alkalinity distributions^{47,48}. The resulting marine carbon cycle has been
450 extensively used and evaluated, including against observations of natural (e.g. $\Delta^{14}\text{C}$) and
451 perturbed anthropogenic carbon cycling. GENIE is also compatible with observational
452 uncertainty⁸ and other (generally higher resolution) carbon cycle model responses to CO₂
453 perturbation⁴⁹⁻⁵¹. The version used here is as summarized by Cao *et al.*⁸ that includes a 76 m
454 deep surface ocean and the cycling of Fe described by Annan and Hargreaves⁵² (except with
455 biological uptake following Doney *et al.*⁵³), but lacks a mixed layer scheme. Our results
456 therefore represent a pessimistic case for weathering and in the real world one might have a
457 slightly shallower surface layer with even higher saturation. The addition of Fe co-limitation
458 of marine biological export results in a <1% change in the projected year 1994 anthropogenic
459 CO₂ inventory compared to the PO₄-only model⁸.

460 We simulated the effects of CO₂ consumption by enhanced weathering on atmospheric
461 CO₂ drawdown and ocean biogeochemistry in two steps. First, we diagnosed the annual CO₂

462 emissions compatible with a particular RCP CO₂ concentration projection over the 21st
463 century by prescribing that CO₂ curve and backing-out emissions. Cross-checking these
464 diagnoses by performing forward simulations with annual CO₂ emissions in the absence of
465 enhanced weathering reproduced the RCP4.5 and RCP8.5 CO₂ curves to within 1 ppm. Then,
466 for each application scenario, we subtracted the annual CO₂ consumption due to enhanced
467 weathering from the diagnosed RCP emissions and forced GENIE with the remainder.

468 The ocean biogeochemistry simulations incorporate reduced atmospheric CO₂ and
469 increases in the alkalinity and dissolved inorganic carbon fluxes. In each case, those for
470 2005–2015 and 2089–2099 are transferred to a 36 × 36 global grid, migrating land fluxes on
471 the weathering model continents to the ocean following standard directional paths. GENIE
472 linearly interpolated these flux forcings for intermediate years. All GENIE runs were based
473 on the same starting state, comprising a 10,000-year pre-industrial spin-up followed by a
474 transient experiment forced by historical changes in atmospheric CO₂ concentration up until
475 year 2006 as described in Cao *et al.*⁸ and Ridgwell *et al.*⁴⁸.

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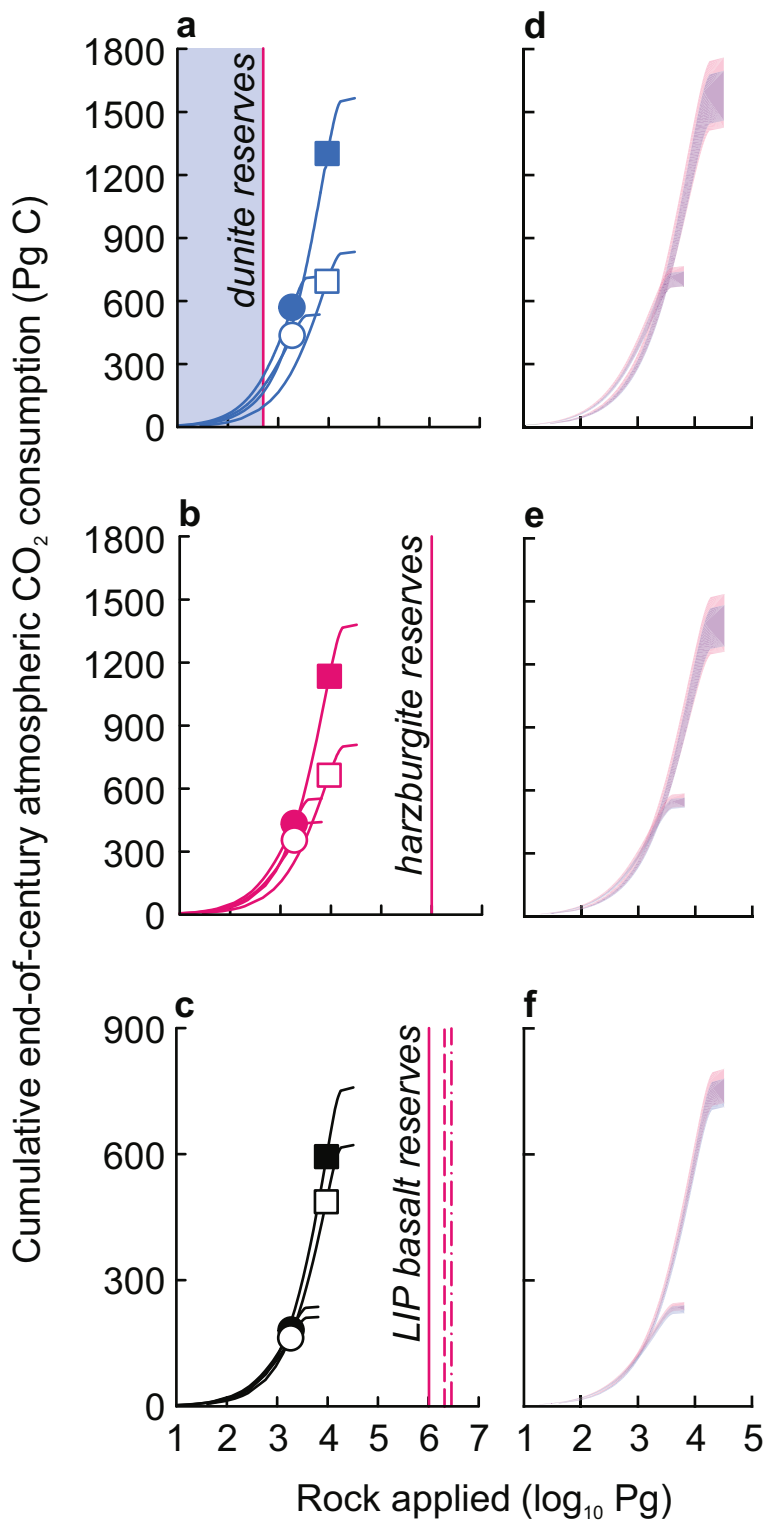
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Table 1. Projected mean global air temperature and change in temperature at year 2100. Values show mean \pm S.D. of five climate models (CMIP5) for the change in end-of-century mean global temperature simulated with the GENIE Earth system model using revised CO₂ trajectories associated with each rock type and application rate with an applied rock mixing depth of 30 cm (Figure 2).

	Warming at 2100 (°C)		Warming averted at 2100 (°C) ^a	
	RCP 4.5	RCP 8.5	RCP 4.5	RCP 8.5
Control (no enhanced weathering) ^b	1.4 \pm 0.01	3.0 \pm 0.01	n/a	n/a
IPCC range of projected warming ^c [Ref.12]	1.1 – 2.6	2.6 – 4.8	n/a	n/a
<i>Enhanced weathering scenario</i>				
Harzburgite (1 kg m ⁻² yr ⁻¹)	0.8 \pm 0.04	2.5 \pm 0.04	0.7 \pm 0.04	0.5 \pm 0.04
Harzburgite (5 kg m ⁻² yr ⁻¹)	-0.77 \pm 0.2	1.4 \pm 0.1	2.2 \pm 0.2	1.6 \pm 0.1
Basalt (1 kg m ⁻² yr ⁻¹)	1.21 \pm 0.02	2.8 \pm 0.01	0.2 \pm 0.02	0.2 \pm 0.01
Basalt (5 kg m ⁻² yr ⁻¹)	0.5 \pm 0.05	2.25 \pm 0.05	0.9 \pm 0.05	0.7 \pm 0.05

^aRelative to control (no enhanced weathering); ^bRelative to 2005; ^cRelative to 1986 – 2005



Application rate:
 squares 5 kg m⁻² yr⁻¹
 circles 1 kg m⁻² yr⁻¹

Mixing depth:
 filled 30 cm
 open 10 cm

RCP:
 purple 4.5
 pink 8.5

