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1 Enhanced weathering strategies for stabilizing climate and averting

2 ocean acidification

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

In 1992, over 170 nations agreed to limit anthropogenic CO₂ emissions to avoid 'dangerous' 33 human-made climate change¹⁶, yet massive expansion of fossil fuel extractions, including 34 shale gas and tar sands, is allowing emissions to grow¹⁷. Avoiding dangerous climate change 35 may therefore require the controversial deployment of Carbon Dioxide Removal (CDR) 36 schemes^{4,18}, so called 'negative emissions' strategies whereby CO₂ is captured and removed 37 from the atmosphere. The Fifth Assessment Report of the Intergovernmental Panel on 38 Climate Change¹² and the U.S. National Research Council Report¹⁸ both recognized enhanced 39 terrestrial weathering of silicate rocks as an important but poorly constrained CDR approach. 40 Currently, natural weathering of silicate and carbonate rocks consumes ~0.25 Pg C yr⁻¹ of 41 atmospheric CO₂, which is ~3% of fossil fuel emissions ¹⁹ (~9–10 Pg C yr⁻¹). Artificially 42 accelerating this land-based CO₂ sink involves the intentional application of pulverised 43 silicate rocks to vegetated landscapes to markedly enhance CO₂ consumption¹⁻⁵. However, 44 45 assessments to date have excluded primary drivers of soil mineral weathering, especially terrestrial ecosystem processes and feedbacks from CO₂ and future climate change, limiting 46 our understanding of its capacity to offset fossil fuel CO₂ emissions¹². 47

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

Our simulations indicate that terrestrial weathering can be markedly increased by distributing pulverised silicate rocks throughout the tropics (30°N to 30°S), potentially consuming hundreds of petagrams (1×10¹⁵ g) of CO₂ by 2100 (Fig. 1). Ensemble median CO₂ consumption by terrestrial weathering increases towards a maximum as the total rock applied increases, with olivine-rich dunite and harzburgite being about twice as effective as basalt for equivalent application rates (Fig. 1a-c). We present CO₂ consumption curves assuming mixing depths of 10 cm and 30 cm for each application rate; 10 cm is likely the minimum mixing depth given intense precipitation events, the distribution of macropores and bioturbation by invertebrates in tropical soils down to depths of 30–50 cm (Supplementary Information). In the model, CO₂ consumption by weathering increases when added rock grains mix deeper in the soil, particularly at the 5 kg m⁻² yr⁻¹ application rate, because mineral saturation, a chemical brake on weathering, occurs more slowly in a larger soil solution volume. Overall CO₂ consumption patterns for a particular RCP scenario show a consistently narrow range of variation across the five ensemble GCMs (Fig. 1d-f). For a given application rate, the magnitude of CO₂ consumption is similar for the business-as-usual (RCP8.5) and medium level mitigation (RCP4.5) scenarios (Fig.1d-f), largely because the runoff for the two scenarios is similar (Supplementary Information).

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

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

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emissions in limiting future warming²⁵.

As CO₂ is removed from the atmosphere by enhancement of the weathering carbon sink, 100 the carbon cycle responds by redistributing carbon among surface reservoirs (atmosphere, 101 ocean, soil, and land biosphere), with CO₂ out-gassing by the ocean in particular offsetting 102 some of the artificial drawdown¹⁷. There is, consequently, a 'rebound' effect whereby each 103 extra mole of CO₂ consumed does not translate into the removal of a mole of atmospheric 104 CO₂ over time. We therefore estimate the effects of our CO₂ consumption fluxes on the 105 RCP4.5 and RCP8.5 atmospheric CO₂ trajectories through the 21st century with the well-106 tested GENIE Earth system model⁸ that broadly captures these responses. Distributing 1kg m⁻ 107 ² yr⁻¹ of pulverised silicates across 20 Mkm² of tropical weathering 'hotspots' lowers 108 109 atmospheric CO₂ concentrations by ~40 ppm (basalt) or ~140 ppm (harzburgite) by year 2100 in both the RCP4.5 and RCP8.5 climate change scenarios (Fig. 2a, b). Increasing the 110 application rate to 5 kg m⁻² yr⁻¹ over the same 20 Mkm² 'hotspot' areas lowers the 111 atmospheric CO₂ concentration further by 150–180 ppm under both RCPs (Fig. 2c, d), with 112 113 an increasing effect at deeper soil mixing depths. For RCP4.5, atmospheric CO₂ by 2100 is reduced from 540 ppm to 390–350 ppm (basalt) or 350–250ppm (harzburgite), sufficient to 114 play a major role in stabilizing climate and avoid seeding long-term amplifying climate 115 feedbacks¹⁷ (Fig. 2). For the business-as-usual RCP8.5 scenario, however, the lowest 116 simulated CO₂ concentration by year 2100 in the high-end weathering scenario is still ~730 117 118 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 close to the target of 350 ppm by 2100, an estimated requirement for restoring planetary 120 energy balance and stabilizing climate¹⁷. 121 122 Future climate warming averted (WA) by engineering CO₂ removal through enhanced weathering is dependent on climate sensitivity and the actual atmospheric CO₂ concentration. 123 124 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 (Table 1). At low application rates, corresponding ranges of WA are 0.2–0.7°C for both 127 RCPs (Table 1). These numbers suggest that, theoretically at least, negative emissions from 128

enhanced weathering could play a role alongside conventional mitigation reducing net CO₂

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

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

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

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

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

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

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

197 Methods

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

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- and A.R. provided model set-up support and advice, M.R.L. analysed the CMIP5 climates. D.J.B. led
- the writing with contributions from all co-authors, especially J.H., A.R., J.Q. and L.L.T.
- 293 **Author Information** Reprints and permissions information is available at www.nature.com/reprints.
- The authors declare no competing financial interests. Correspondence and requests for materials
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Figure Legends

297	Figure 1. Enhanced weathering from pulverised silicate rock additions to the tropics
298	increases CO ₂ consumption. End-of-century CO ₂ 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 CO ₂ consumption and total rock applied when application is limited to 20 Mkm ²
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 CO ₂ with projected 21 st century
313	climate change. Effects of low additions (1 kg m ⁻² yr ⁻¹) of silicate rock to 20 Mkm ² of
314	tropical weathering hotspots for two mixing depths (10 cm and 30 cm) on the atmospheric
315	CO ₂ 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 CO ₂ . Effects of increased alkalinity fluxes				
323	resulting from additions of 1 kg m ⁻² yr ⁻¹ of silicate rock to 20 Mkm ² 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.				

Methods

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

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

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

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

$$Rate_{m} = SA_{m} \sum_{i} \left[k_{i,m}^{298.15} exp \left[\frac{-E_{i,m}}{R} \left(\frac{1}{T} - \frac{1}{298.15} \right) \right] a_{i}^{n_{i,m}} \left(1 - \left[\frac{Q_{m}}{Ksp_{m}} \right] \right) \right] \quad \text{Eq. (1)}$$

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

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

Validation. Simulated terrestrial fluxes of CO_2 consumed by rock weathering in 42 watersheds worldwide³⁹ using the CRU-3 climate⁴⁰ at $1^{\circ} \times 1^{\circ}$ resolution are validated against fluxes derived from catchment-scale estimates based on stream-water chemistry

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

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

Geoengineering simulations. For the atmospheric CO₂ concentration trajectories defined by the two RCPs considered here (RCP4.5 and RCP8.5), the five General Circulation Models (GCMs) produce monthly temperature, relative humidity and precipitation to drive SDGVM. Monthly climate datapoints closest to the desired coordinates are bilinearly interpolated in space before daily values for the month are estimated using climate statistics. These estimated daily climates force SDGVM. Distributed silicate rock grains are treated as perfect monomineralogical spheres with a nominal starting diameter of 10 μm. Initial total surface areas for the added silicates are calculated for each mineral using the total mass applied, specified diameter, weight fraction for the mineral, mineral specific gravity and the equations for the volume and surface area of a sphere. As weathering progresses, mass is removed and the fractional change in total surface area is estimated using the fractional change in mass raised to the power ¾3. This treatment assumes that each particle is a shrinking sphere. The total mass and surface area for each mineral are increased according to dose rate. No attempt is made to model a particle size distribution for either the starting silicate rock grain size or 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 primary soil minerals are corrected for erosion and relief⁷. The model assumes no change in 433 porosity or water movement with depth, and there is no preferential transport of different 434 particle sizes. 435 Mineralogy of pulverized silicates. The mineralogy of each simulated pulverized silicate 436 rock is listed in Table S4. The model basalt silicate mineralogy is based on the normative 437 composition for a normal alkali tholeiitic basalt⁴⁴, neglecting some minor phases such as 438 magnetite. Our dunite composition follows Kogel et al. 45. We use the mineralogy of the 439 Troodos harzburgite, with lizardite rather than chrysotile (asbestos) as this is the dominant 440 serpentine near Troodos⁴⁶ and as it is sensible to avoid rocks with a large proportion of 441 asbestos for health reasons. Our results are therefore conservative with respect to the 442 proportion of unserpentinised olivine and the relative amounts of lizardite and chrysotile 443 444 present. 445 GENIE Earth system global CO₂ and ocean biogeochemistry modelling. The climate and ocean circulation of the GENIE Earth system model has been calibrated by 2-D reanalysis 446 447 fields of surface air temperature and humidity and 3-D observational fields of ocean distributions of temperature and salinity⁴⁷. The carbon cycle is calibrated against observed 448 ocean phosphate and alkalinity distributions 47,48. The resulting marine carbon cycle has been 449 extensively used and evaluated, including against observations of natural (e.g. Δ^{14} C) and 450 perturbed anthropogenic carbon cycling. GENIE is also compatible with observational 451 uncertainty⁸ and other (generally higher resolution) carbon cycle model responses to CO₂ 452 perturbation ⁴⁹⁻⁵¹. The version used here is as summarized by Cao et al. 8 that includes a 76 m 453 deep surface ocean and the cycling of Fe described by Annan and Hargreaves⁵² (except with 454 biological uptake following Doney et al. 53), but lacks a mixed layer scheme. Our results 455 456 therefore represent a pessimistic case for weathering and in the real world one might have a slightly shallower surface layer with even higher saturation. The addition of Fe co-limitation 457 458 of marine biological export results in a <1% change in the projected year 1994 anthropogenic CO₂ inventory compared to the PO₄-only model⁸. 459 We simulated the effects of CO₂ consumption by enhanced weathering on atmospheric 460 CO₂ drawdown and ocean biogeochemistry in two steps. First, we diagnosed the annual CO₂ 461

- emissions compatible with a particular RCP CO₂ concentration projection over the 21st
 century by prescribing that CO₂ curve and backing-out emissions. Cross-checking these
 diagnoses by performing forward simulations with annual CO₂ emissions in the absence of
 enhanced weathering reproduced the RCP4.5 and RCP8.5 CO₂ curves to within 1 ppm. Then,
 for each application scenario, we subtracted the annual CO₂ consumption due to enhanced
 weathering from the diagnosed RCP emissions and forced GENIE with the remainder.
- The ocean biogeochemistry simulations incorporate reduced atmospheric CO₂ and 468 469 increases in the alkalinity and dissolved inorganic carbon fluxes. In each case, those for 2005–2015 and 2089–2099 are transferred to a 36 × 36 global grid, migrating land fluxes on 470 471 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 year 2006 as described in Cao et al. 8 and Ridgwell et al. 48. 475

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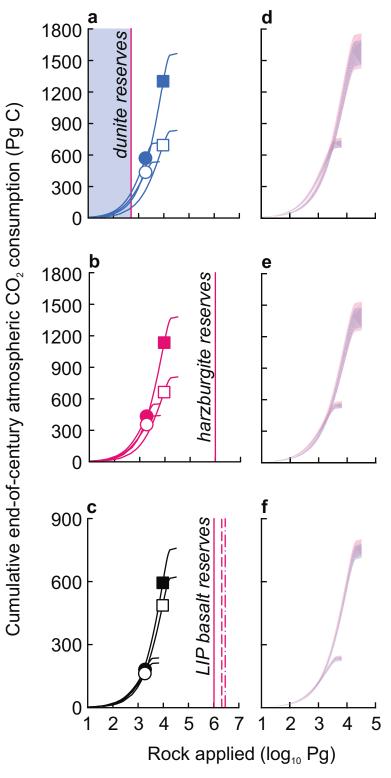
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Table 1. Projected mean global air temperature and change in temperature at year 2100. Values show mean \pm S.D. of five climate models (CMIP5) for the change in end-of-century mean global temperature simulated with the GENIE Earth system model using 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 ± 0.01	3.0 ± 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
Enhanced weathering scenario				
Harzburgite (1 kg m ⁻² yr ⁻¹)	0.8 ± 0.04	2.5 ± 0.04	0.7 ± 0.04	0.5 ± 0.04
Harzburgite (5 kg m ⁻² yr ⁻¹)	-0.77 ± 0.2	1.4 ± 0.1	2.2 ± 0.2	1.6 ± 0.1
Basalt (1 kg m ⁻² yr ⁻¹)	1.21 ± 0.02	2.8 ± 0.01	0.2 ± 0.02	0.2 ± 0.01
Basalt (5 kg m ⁻² yr ⁻¹)	0.5 ± 0.05	2.25 ± 0.05	0.9 ± 0.05	0.7 ± 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



