

## **Supplementary Information for**

New estimates of the storage permanence and ocean co-benefits of enhanced rock

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## 52 Supplementary Information Text

53 Earth system model. We explore the impacts of large-scale CDR using a 'carbon-centric' 54 version of the Grid Enabled Integrated Earth system model — cGENIE. The ocean physics and 55 climate model components of cGENIE comprise a reduced physics (frictional geostrophic) 3-D ocean circulation model coupled to a 2-D energy-moisture balance model (EMBM) and a 56 57 dynamic-thermodynamic sea ice model [1]. Heat, salinity, and biogeochemical tracers are 58 transported via parameterized isoneutral diffusion and eddy-induced advection [2]. The ocean 59 model exchanges heat and moisture with the atmosphere, sea ice, and land while being forced at 60 the ocean surface by zonal and meridional wind stress according to a specified static wind field. 61 Heat and moisture are horizontally mixed throughout the atmosphere and exchange heat and 62 moisture with the ocean and land surfaces, with precipitation occurring above a given relative 63 humidity threshold. The sea ice model tracks horizontal ice transport and exchanges of heat and 64 fresh water, using the thickness, areal fraction, and concentration of ice as prognostic variables. Full descriptions of the climate model and ocean physics can be found in [1, 2]. The ocean model 65 is configured here as a 36 x 36 equal-area grid (uniform in longitude and sine of latitude) with 66 67 16 logarithmically spaced depth levels and seasonal forcing at the ocean surface.

68 The ocean and sediment biogeochemistry modules in cGENIE control air-sea gas exchange, the 69 transformation and repartitioning of biogeochemical tracers within the ocean, and the impacts of 70 shallow sediment diagenesis on calcium carbonate formation/dissolution and burial. The ocean 71 biological carbon pump is driven by a parameterized uptake rate of nutrients in the surface ocean, 72 with this flux converted stoichiometrically to biomass that is then partitioned into particulate or 73 dissolved organic matter for downstream advective transport, sinking, and remineralization 74 within the ocean interior. Dissolved organic matter is transported with the ocean circulation and 75 decays according to a specified time constant, while particulate organic matter is instantaneously 76 exported from the surface ocean and is remineralized within the ocean interior following an 77 exponential decay function with a specified remineralization length scale. The ocean 78 biogeochemistry also contains a fully coupled carbonate system, which tracks individual 79 dissolved inorganic carbon (DIC) species, dissolved alkalinity, and ocean pH. Calcium carbonate 80 forms in surface ocean grid cells at a stoichiometric ratio with organic matter production (the so-81 called "rain ratio") and is exported as a solid species and is dissolved in the ocean interior or 82 shallow marine sediments depending on ambient temperature, pressure, and carbonate chemistry 83 [3, 4]. A simple scheme for shallow sediment diagenesis allows us to run the ocean alkalinity 84 cycle as an open system, with delivery from weathering of the land surface and ultimate burial 85 as calcite (CaCO<sub>3</sub>) in marine sediments). More detailed description and validation of the ocean 86 and sediment biogeochemistry in cGENIE is provided in [5, 6].

87 **Terrestrial carbon exchange.** We implement a simple model of carbon exchange with the 88 terrestrial biosphere in which aboveground biomass (vegetation) and soil carbon are treated as 89 global pools that respond to temperature and atmospheric  $pCO_2$  (e.g., a "slab" or "box" terrestrial 90 biosphere). The model tracks changes in the size of the aboveground carbon reservoir 91 (vegetation, V) and soil carbon (S) according to:

92 
$$\frac{dV(t)}{dt} = N(t) - L(t)$$
, [Eq. S1]

93 
$$\frac{dS(t)}{dt} = L(t) - R(t)$$
, [Eq. S2]

Where N(t) represents net primary production (GtC y<sup>-1</sup>), L(t) represents the production rate of litterfall (GtC y<sup>-1</sup>), and R(t) represents soil respiration (GtC y<sup>-1</sup>). Net primary production is parameterized as a function of atmospheric  $pCO_2$  according to:

97 
$$N(t) = N_0 \left[ 1 + B \ln \left( \frac{C(t)}{C_0} \right) \right], \qquad [Eq. S3]$$

98 Where C(t) is atmospheric  $pCO_2$ ,  $N_0$  is net primary production at a baseline atmospheric  $pCO_2$ 99 ( $C_0$ ), and B is a growth rate parameter. The rate of litterfall production is given by:

100 
$$L(t) = V(t) \left[ \Lambda_{veg} V(t) + \Lambda_0 \right]^{-1} , \qquad [Eq. S4]$$

101 Where the  $\Lambda_{veg}$  and  $\Lambda_{\theta}$  terms describe an intrinsic turnover time for vegetation (y). Soil 102 respiration is parameterized as a function of temperature according to:

103 
$$R(t) = \Gamma S(t) Q_{10}^{\frac{T(t) - T_0}{10}}$$
, [Eq. S5]

104 Where  $\Gamma$  is the annual soil carbon turnover rate at reference temperature  $T_0$  (y<sup>-1</sup>) and  $Q_{10}$ 105 represents a parameter describing the factor change in soil respiration rate for a 10°C change in 106 temperature.

107 Once  $C_0$  and  $T_0$  are defined, the model contains six parameters ( $N_0$ , B,  $\Lambda_{veg}$ ,  $\Lambda_0$ ,  $\Gamma$ , and  $Q_{10}$ ). We 108 use a stochastic approach to account for uncertainty in the slab biosphere parameterization. First, 109 we randomly generate  $2 \times 10^6$  parameter sets from the ranges given in Table S2. These parameter 110 sets are implemented in a stand-alone (offline) version of the slab biosphere model driven by 111 temperature and pCO<sub>2</sub> trajectories from Representative Concentration Pathway (RCP) and 112 Extended Concentration Pathway (ECP) scenarios [7]. These parameter sets are then filtered for 113 those that yield results consistent with modern observations of soil carbon stocks, aboveground 114 net primary production, and vegetation turnover time (Fig. S2) and that result in a dynamic 115 response that falls within the range of CMIP5 projections for changes in vegetation, soil, and 116 total land organic carbon pools to the end of the century (Fig. S3-S5), yielding a filtered ensemble 117 of n = 7,551 parameter sets. A subset (n = 3,000) of these parameter sets were then implemented 118 in a set simulations in which the slab biosphere is fully coupled to cGENIE and are again filtered 119 based on modern observations and end-of-century projections to yield our final ensemble of n =120 980 Earth system model simulations.

121 Model spinup, control simulations, and CDR scenarios. The model climate system and ocean 122 carbonate/alkalinity cycle are spun up to steady state using a two-stage procedure. First, the 123 model is run as a closed system for 20 kyr with atmospheric abundances of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O 124 imposed at preindustrial values to bring the ocean-atmosphere system and shallow sediments 125 into steady state. This run is used to diagnose the approximate steady state burial flux of calcium 126 carbonate in marine sediments, which is then imposed as a weathering flux of calcium and 127 alkalinity in a second stage spinup in which the ocean and sediments are allowed to evolve as an 128 open system. The second stage spinup is run for 75 kyr to allow the ocean alkalinity budget to 129 achieve steady state.

All subsequent simulations are branched from the open system spinup at model year 1765 and run to year 2300 according to the Representative Concentration Pathway (RCP) and Extended 132 Concentration Pathway (ECP) scenarios for atmospheric  $CO_2$ ,  $CH_4$ , and  $N_2O$  [7]. Time-varying 133 atmospheric abundances of  $CH_4$  and  $N_2O$  are imposed according to a given RCP/ECP trajectory 134 for all simulations, while atmospheric  $CO_2$  abundance is emission-driven. The emission 135 trajectory for a given RCP is first computed by the model by prescribing the atmospheric  $CO_2$ 136 trajectory for that scenario, with all subsequent runs utilizing the emission trajectory diagnosed 137 in cGENIE for each RCP/ECP pathway.

Our simulations of carbon dioxide capture are designed to represent two distinct CDR deployment modes. In simulations of CDR via enhanced rock weathering (ERW), we specify an initial capture rate (in  $GtCO_2 y^{-1}$ ) which is then translated into a removal of  $CO_2$  from the atmosphere and a corresponding flux of dissolved inorganic carbon (DIC) and alkalinity to the coastal ocean. We simulate ERW using two distinct feedstocks, with idealized stoichiometries for natural silicate and carbonate minerals:

144 
$$CaSiO_3 + 2CO_2 + 3H_2O \rightarrow Ca^{2+} + 2HCO_3^{-} + H_4SiO_4$$
, [Eq. S6]

145 
$$CaCO_3 + CO_2 + H_2O \rightarrow Ca^{2+} + 2HCO_3^-$$
 [Eq. S7]

Fluxes of DIC and alkalinity to the ocean per ton of CO<sub>2</sub> removed are controlled according to the stoichiometries shown, and are routed to the ocean according to simulated topological network (STN) data [8]. Note that on arbitrarily long timescales the formation and burial of carbonate minerals in marine sediments (e.g., the reverse of carbonate dissolution) releases CO<sub>2</sub>, undoing most or all of the capture associated with ERW using carbonate rock and around half of the capture associated with ERW using silicate feedstock.

152 We also simulate an alternative carbon cycle intervention meant to represent direct CDR or 153 mitigation of emissions in excess of that implied by a given RCP pathway, in which we reduce 154 CO<sub>2</sub> emission rates globally by a specified value relative to the control emission rates for a given 155 RCP. The key difference between this idealized style of CDR deployment and those associated 156 with ERW is that it is assumed that the carbon is instantaneously and permanently removed from 157 the surface system rather than being transiently repartitioned into a non-radiative but potentially 158 labile form of surface carbon (some of which will ultimately be removed from Earth's surface). 159 This style of intervention could be viewed as consistent with a variety of distinct strategies but 160 is most directly analogized to direct air capture and storage (DACS) or emissions mitigation. We explore a wide range of CDR deployment scales between 0.5 - 40 GtCO<sub>2</sub> y<sup>-1</sup>, a range meant to 161 162 be inclusive of relatively modest CDR deployment on the low end to a deployment scale 163 sufficient to offset most or all of global anthropogenic CO<sub>2</sub> emissions on the other.

164 **Theoretical formulation of carbon leakage.** Carbon capture and leakage through NETs can be 165 defined based on fluxes between the major carbon reservoirs at Earth's surface relative to a 166 deployed carbon cycle intervention, represented here as  $J_{CDR}$  but meant to incapsulate both direct 167 carbon dioxide removal and mitigated fossil fuel emissions. Below we develop a basic 168 framework for CO<sub>2</sub> capture efficiency as well as carbon leakage resulting from anthropogenic 169 carbon cycle intervention.

170 The total mass of carbon that is transferrable on anthropogenic timescales,  $C_{all}$  (mol), can be 171 defined as the summed mass of carbon in three reservoirs: $\approx$ 

172 
$$C_{all} = C_{atm} + C_{ocn} + C_{land}$$
, [Eq. S8]  
173

where  $C_{atm}$ ,  $C_{ocn}$  and  $C_{land}$  represent the carbon reservoir size (mol) of the atmosphere, ocean, and land, respectively. Time-integrated flows of carbon caused by CDR deployment (or mitigated emissions) can be defined based on the difference in reservoir size between a given intervention experiment (*exp*) and a corresponding control simulation (*ctrl*):

178 
$$\Delta C_X = C_X^{exp} - C_X^{ctrl} , \qquad [Eq. S9]$$

179 where X = all, *atm*, *ocn* or *land*. Given that carbon mass balance must be maintained with or 180 without CDR/mitigated emissions:

181 
$$\Delta C_{all} = \Delta C_{atm} + \Delta C_{ocn} + \Delta C_{land} \quad .$$
 [Eq. S10]

182 We can then define capture efficiency with reference to the long-term impact on the atmospheric183 carbon reservoir as:

184 
$$\eta_{CDR} = \frac{-\Delta C_{atm}}{\int_{2030}^{t} J_{CDR} dt'}$$
, [Eq. S11]

- 185 We can also explicitly link the cumulative impact of CDR or mitigated emissions on the mass
- 186 of atmospheric carbon with a corresponding alkalinity change in the ocean ( $\Delta A_{ocn}$ , mol)
- 187 associated with direct removal or mitigated emissions (base) and CDR through enhanced silicate
- 188 weathering (ESW) or enhanced carbonate weathering (ECW).

189 
$$\Delta A_{ocn} = \begin{cases} 0 & (CDR = base) \\ \int_{2030}^{t} J_{CDR} dt' & (CDR = ESW) \\ 2 \int_{2030}^{t} J_{CDR} dt' & (CDR = ECW) \end{cases},$$
[Eq. S12]

We can also relate the time-integrated change in total Earth surface carbon mass to cumulative
 CO<sub>2</sub> removal or mitigated emissions across the same range of intervention styles:

192 
$$\Delta C_{all} = \begin{cases} -\int_{2030}^{t} J_{CDR} dt' & (CDR = base) \\ 0 & (CDR = ESW) \\ \int_{2030}^{t} J_{CDR} dt' & (CDR = ECW) \end{cases}$$
 [Eq. S13]

193 Note that Eq. S13 assumes timescales less than 10<sup>5</sup> years. Meanwhile, the land and ocean carbon 194 reservoirs do not exchange directly but only through the atmosphere reservoir. Thus:

195 
$$\Delta C_{land} = -\int_{2030}^{t} (J_{land-air}^{exp} - J_{land-air}^{ctrl}) dt', \qquad [Eq. S14]$$

196 
$$\Delta C_{ocn} = \Delta A_{ocn} - \int_{2030}^{t} (J_{sea-air}^{exp} - J_{sea-air}^{ctrl}) dt' . \qquad [Eq. S15]$$

197 Using Eqs. S11-S15, we can express capture efficiency with respect to carbon fluxes:

198 
$$\eta_{CDR} = \frac{\int_{2030}^{t} \left[ J_{CDR} - (J_{sea-air}^{exp} - J_{sea-air}^{ctrl}) - (J_{land-air}^{exp} - J_{land-air}^{ctrl}) \right] dt'}{\int_{2030}^{t} J_{CDR} dt'} \quad [Eq. S16]$$

199 Carbon leakage can then be defined as:

200 
$$p_{CDR} \equiv 1 - \eta_{CDR} = \frac{\int_{2030}^{t} \left[ (J_{sea-air}^{exp} - J_{sea-air}^{ctrl}) + (J_{land-air}^{exp} - J_{land-air}^{ctrl}) \right] dt'}{\int_{2030}^{t} J_{CDR} dt'}$$
, [Eq. S17]  
201

- 202 We define a baseline "Earth system leakage" (CDR = base in the expressions above) which does
- 203 not involve any direct impacts on ocean chemistry. Then additional carbon leakage during ERW, 204  $\Delta p_{ERW}$ , can be defined using Eqs. S16-S17:

205 
$$\Delta p_{ERW} = \begin{cases} p_{ESW} - p_{base} = 1 - \frac{\Delta C_{ocn}}{\Delta A_{ocn}} \left(1 + \frac{\Delta C_{land}}{\Delta C_{ocn}}\right) & (ERW = ESW) \\ p_{ECW} - p_{base} = 2 \left[1 - \frac{\Delta C_{ocn}}{\Delta A_{ocn}} \left(1 + \frac{\Delta C_{land}}{\Delta C_{ocn}}\right)\right] & (ERW = ECW) \end{cases},$$
 [Eq. S18]

where  $\Delta'$  indicates the difference from a corresponding baseline (direct removal or mitigated emissions) scenario:

$$\Delta' C_X = C_X^{ERW} - C_X^{base} . \qquad [Eq. S19]$$

Here, ERW can denote either ESW or ECW. Our ensemble of simulations with stochastic slab biosphere parameterization suggests  $\Delta' C_{land} / \Delta' C_{ocn} < 0.05$  and thus we can assume  $\Delta' C_{ocn} >>$  $\Delta' C_{land}$ . Therefore, regardless of the response of the terrestrial biosphere we can simplify the expression for residual carbon leakage during ERW to:

213 
$$\Delta p_{ERW} = \begin{cases} 1 - \frac{\Delta' C_{ocn}}{\Delta' A_{ocn}} & (ERW = ESW) \\ 2 \left[ 1 - \frac{\Delta' C_{ocn}}{\Delta' A_{ocn}} \right] & (ERW = ECW) \end{cases}, \qquad [Eq. S20]$$

Using the time-integrated global carbon flux and reservoir shifts from our ensemble of simulations and Eq. (S20) results in:

216 
$$\frac{\Delta' C_{ocn}}{\Delta' A_{ocn}} \approx 0.9$$
, [Eq. S21]

217 
$$\Delta p_{ECW} \approx 2\Delta p_{ESW}$$
, [Eq. S22]

This is consistent with the near-term equilibrium values for residual carbon leakage given in Main Text Fig. 2. In both cases the carbon storage efficiency is greater than that conventionally assumed [9, 10], which we suggest results from transport of carbon into the ocean interior.



Figure S1. Surface ocean carbonate chemistry parameters in historical cGENIE simulation compared to gridded observational data. Shown at left are year 2000 results for concentrations of dissolved inorganic carbon ([DIC]), alkalinity ([ALK]), aragonite saturation state ( $\Omega_{aragonite}$ ), and pH. Shown on the right are observational data from the GLODAPv2 dataset [11].



266 Figure S2. Comparison of Earth system model results with modern empirical observations of soil organic carbon (soil C), aboveground net primary productivity (NPP), and vegetation turnover time. Bars show modern observations [12], while crosses and open circles show Earth system model results using default slab biosphere parameters and the range for our filtered ensemble, respectively. 





Figure S3. Comparison of Earth system model results with CMIP5 model range for aboveground vegetation carbon through the next century. Shaded envelope shows large-ensemble results from our intermediate complexity Earth system model (n = 980), while solid lines show individual members of the CMIP5 ensemble (https://esgf-node.llnl.gov/search/cmip5/).

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324 carbon through the next century. Shaded envelope shows large-ensemble results from our intermediate complexity Earth system model (n = 980), while solid lines show individual members 328 329 of the CMIP5 ensemble (https://esgf-node.llnl.gov/search/cmip5/).

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- 336



344 surface carbon through the next century. Shaded envelope shows large-ensemble results from our intermediate complexity Earth system model (n = 980), while solid lines show individual members of the CMIP5 ensemble (https://esgf-node.llnl.gov/search/cmip5/). 

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**Figure S6.** Atmospheric carbon dioxide ( $pCO_2$ ), global average warming since the preindustrial period ( $\Delta T$ ), and average surface ocean aragonite saturation state ( $\Omega_{arg}$ ) for a range of emission and CDR scenarios. Results are shown for four Representative Concentration Pathway (RCP) scenarios (left-right), and for control runs with no CDR (black), direct air capture and storage and/or additional mitigated emissions (grey), enhanced silicate weathering (ESW; green), and enhanced carbonate weathering (ECW; blue). All CDR scenarios are for a 10 GtCO<sub>2</sub> y<sup>-1</sup> deployment beginning in 2030. Values for  $\Delta T$  are calculated relative to the period 1850-1900.





**Figure S7.** Carbon leakage through 2100 across a range of emission and carbon dioxide removal (CDR) scenarios, shown as a time-integrated percentage relative to CDR deployment level. Black curves and error envelopes show the ensemble median and uncertainty on baseline (modulated emissions) response, while red and yellow curves and error envelopes show ensemble median and uncertainty for the residual enhanced carbonate weathering (ECW) and enhanced silicate weathering (ESW), respectively, after correcting to the baseline response.





407 Figure S8. Carbon leakage as a function of deployment level of carbon dioxide removal (CDR) or additional mitigated emissions. Results are shown for each Representative Concentration Pathway (RCP) scenario in model years 2070 and 2100, for baseline (base) intervention, enhanced silicate weathering (ESW) and enhanced carbonate weathering (ECW).

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Figure S9. Ocean carbon cycle response to carbon dioxide removal in our Earth system model for RCP2.6. Shown at left are sea-air CO<sub>2</sub> fluxes (top), depth-integrated inventory of dissolved inorganic carbon ([DIC]int; middle), and zonally averaged DIC concentrations (bottom) for the baseline (modulated emissions) case and the enhanced silicate weathering (ESW) scenario. Shown at right are anomaly plots of sea-air flux (top), depth-integrated DIC inventory (middle), and zonally averaged DIC (bottom) between the ESW scenario and the equivalent modulated emissions case. Results are shown for year 2070 and a continuous CDR deployment level of 10 GtCO<sub>2</sub> y<sup>-1</sup> starting in 2030. Note that DIC concentration/anomaly results (D-I) are shown excluding the uppermost grid cell (80m).



Figure S10. Ocean carbon cycle response to carbon dioxide removal in our Earth system model for RCP6.0. Shown at left are sea-air CO<sub>2</sub> fluxes (top), depth-integrated inventory of dissolved inorganic carbon ([DIC]int; middle), and zonally averaged DIC concentrations (bottom) for the baseline (modulated emissions) case and the enhanced silicate weathering (ESW) scenario. Shown at right are anomaly plots of sea-air flux (top), depth-integrated DIC inventory (middle), and zonally averaged DIC (bottom) between the ESW scenario and the equivalent modulated emissions case. Results are shown for year 2070 and a continuous CDR deployment level of 10 GtCO<sub>2</sub> y<sup>-1</sup> starting in 2030. Note that DIC concentration/anomaly results (D-I) are shown excluding the uppermost grid cell (80m).



Figure S11. Ocean carbon cycle response to carbon dioxide removal in our Earth system model for RCP8.5. Shown at left are sea-air CO<sub>2</sub> fluxes (top), depth-integrated inventory of dissolved inorganic carbon ([DIC]<sub>int</sub>; middle), and zonally averaged DIC concentrations (bottom) for the baseline (modulated emissions) case and the enhanced silicate weathering (ESW) scenario. Shown at right are anomaly plots of sea-air flux (top), depth-integrated DIC inventory (middle), and zonally averaged DIC (bottom) between the ESW scenario and the equivalent modulated emissions case. Results are shown for year 2070 and a continuous CDR deployment level of 10 GtCO<sub>2</sub> y<sup>-1</sup> starting in 2030. Note that DIC concentration/anomaly results (D-I) are shown excluding the uppermost grid cell (80m).





**Figure S12.** Global average surface ocean aragonite saturation state ( $\Omega_{arg}$ ) as a function of 509 deployment level of carbon dioxide removal (CDR) or additional mitigated emissions. Results are 510 shown for each Representative Concentration Pathway (RCP) scenario in model years 2070 and 511 2100, for baseline (base) intervention, enhanced silicate weathering (ESW) and enhanced 512 carbonate weathering (ECW).

Parameter (symbol [units])	Equation	
Vegetation (V [GtC])	$\frac{dV(t)}{dt} = N(t) - L(t)$	
Soil carbon (S [GtC])	$\frac{dS(t)}{dt} = L(t) - R(t)$	
Net primary production ( $N$ [GtC y <sup>-1</sup> ]) <sup>a</sup>	$N(t) = N_0 \left[ 1 + \beta \ln \left\{ \frac{p \text{CO}_2(t)}{p \text{CO}_2^{ref}} \right\} \right]$	
Litterfall ( <i>L</i> [GtC y <sup>-1</sup> ])	$L(t) = \frac{V(t)}{\Lambda}$	
Respiration ( <i>R</i> [GtC y <sup>-1</sup> ]) <sup>b</sup>	$R(t) = \Gamma S(t) Q_{10}^{\frac{T(t)-T_0}{10}}$	
Turn over $(\Lambda [y])$	$\Lambda = aV(t) + b$	

#### Table S1. Governing equations for the terrestrial-biosphere box module

<sup>a</sup> pCO<sub>2</sub> is in units of ppm and pCO<sub>2</sub><sup>ref</sup> is a reference pCO<sub>2</sub> (365 ppm). <sup>b</sup> T is the global average land surface temperature excluding Antarctica and Greenland (°C) and  $T_0$  is a reference T (15 533 534 °C).

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571	Table S2. Parameter ranges examined and values chosen for the terrestrial-biosphere box
572	module (Table S1).

Parameter [units]	Examined plausible range <sup>a</sup>	Default value <sup>b</sup>
$N_0$ [GtC y <sup>-1</sup> ]	60 - 80	60
$\beta$ [dimensionless]	0.3 - 0.9	0.42
Г [у-1]	0.04 - 0.07	0.04
$Q_{10}$ [dimensionless]	1.0 - 5.0	2.6
$a [y \text{ GtC}^{-1}]$	-0.02 - 0.004	-0.0056
<i>b</i> [y]	5 - 18	18

<sup>a</sup> Ranges cover values reported in [13]. <sup>b</sup> The combination of the parameter values listed in the third column is adopted as default, which yields estimates consistent with observations and CMIP5 forecasts.

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