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importance of terrestrial weathering changes The multimillennial recovery of the global carbon cycle: a two-2 dimensional perspective 3 4 5 Marc-Olivier Brault^{1,*}, H. Damon Matthews², and Lawrence A. Mysak³ 7 8 ¹Department of Geography, McGill University, Montreal, Canada 9 ²Department of Geography, Planning and Environment, Concordia University, Montreal, Canada 10 ³Department of Atmospheric and Oceanic Sciences, McGill University, Montreal, Canada 11 12 13 14 15 16 *Corresponding Author: Marc-Olivier Brault, Department of Geography, McGill University Montreal, Canada H3A 0B9. E-mail: marc-olivier.brault@mail.mcgill.ca 17

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1 Abstract

2 In this paper, we describe the development and application of a new spatially-explicit weathering

scheme within the University of Victoria Earth System Climate Model (UVic ESCM). We

integrated a dataset of modern-day lithology with a number of previously devised

5 parameterizations for weathering dependency on temperature, primary productivity, and runoff.

6 We tested the model with simulations of future carbon cycle perturbations, comparing a number

of emission scenarios and model versions with each other and with zero-dimensional equivalents

of each experiment. Overall, we found that our two-dimensional weathering model versions

were more efficient in restoring the carbon cycle to its pre-industrial state following the pulse

emissions than their zero-dimensional counterparts; however, in either case the effect of this

weathering negative feedback on the global carbon cycle was small on timescales of less than

1000 years. According to model results, the largest contribution to future changes in weathering

rates came from the expansion of tropical and mid-latitude vegetation in grid cells dominated by

weathering-vulnerable rock types, whereas changes in temperature and river runoff had a more

modest direct effect. Our results also confirmed that silicate weathering is the only mechanism

that can lead to a full recovery of the carbon cycle to pre-industrial levels on multi-millennial

17 timescales.

Keywords

20 Weathering; Carbon cycle modeling; Future climate change; Biogeochemistry.

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1 1 Introduction

2 1.1 Rationale

3 The weathering of carbonate and silicate rocks on land is a key process in the global carbon

4 cycle and, through its coupling with calcium carbonate deposition in the ocean, is the primary

5 sink of carbon on geologic timescales (Urey, 1952; Walker et al., 1981). The rate at which these

processes remove carbon from the Earth system is sensitive to changes in the environment,

notably temperature (Berner, 1991), biological productivity (Lenton and Britton, 2006) and

8 perhaps more indirectly, river runoff (Walker and Kasting, 1992). This gives rise to a negative

9 feedback mechanism which regulates the global climate on multimillennial time scales.

10 However, there have been but very few quantitative assessments of its impacts on carbon cycling

and ocean biogeochemistry, and its relevance over time frames of 10⁴ years or shorter is largely

12 unknown.

Here, we introduce a new model of rock weathering developed for use within the University of

Victoria Earth System Climate Model (UVic ESCM); this model incorporates a spatially explicit

interactive computation of weathering rates to close the global carbon cycle on multi-millennial

time scales. The model is based on a lithology-dependent calculation of steady-state weathering

fluxes, which are modulated by transient changes in environmental conditions akin to the 0-D

carbon cycle models already present in the literature (e.g. Meissner et al., 2012). We tested the

19 model with simulations of future climate changes following anthropogenic carbon emissions,

20 comparing the output to that of earlier weathering models, both 2-D (Colbourn et al., 2013) and

21 0-D (Lenton and Britton, 2006).

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1.2 The rock weathering cycle

2 The chemical weathering of rocks is characterized by the cleavage of bonds of the mineral lattice

3 by water, often in the presence of a secondary weathering agent – hydronium or OH ions, low

4 molecular weight organic chelators, or carbonic acid (H₂CO₃; a product of carbon dioxide

5 dissolution in rainwater). Rock weathering products, including calcium and bicarbonate ions

6 (respectively the most abundant cation and anion in most river waters), can be carried away with

7 runoff to rivers and into the ocean. For example, calcium carbonate dissolution by carbonic acid

8 is given by (Archer et al., 1997):

9
$$CaCO_3 + CO_2 + H_2O \rightarrow Ca^{2+} + 2HCO_3^-$$
 (1)

The influx of dissolved inorganic carbon (henceforth DIC) and alkalinity to the ocean surface layer is balanced by the precipitation and burial of biogenic calcium carbonate (CaCO₃) in the marine sediments, and ocean alkalinity is a key factor in determining the carbonate compensation depth (CCD), the depth below which the dissolution rate of calcium carbonate exceeds its precipitation rate. In the long term this allows the ocean to maintain a remarkably stable alkalinity, as any increases in ocean acidity (such as can be caused by a CO₂ invasion from the atmosphere) can be neutralized by elevating the CCD, which dissolves carbonate sediments and releases carbonate ions (CO₃²⁻) back into the ocean. This oceanic buffer factor, along with carbonate dissolution on land (due to weathering), is the primary means through which ocean alkalinity is restored, and is responsible for maintaining both atmospheric and oceanic pCO₂ close to equilibrium. In short, the weathering of calcium carbonate can accelerate the transfer of CO₂ between the atmosphere and ocean, but does not contribute to a permanent return of carbon to the geologic reservoir (Ridgwell and Zeebe, 2005; Sarmiento and Gruber, 2006).

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- 1 A certain fraction of rock weathering reactions involve a weakening of chemical bonds in the
- 2 mineral lattice on contact with water whereby hydrogen ions replace positively charged cations
- 3 (mostly Ca²⁺ and Mg²⁺) which are bounded to negatively charged ions, most particularly SiO₄
- 4 (silicate) structures. One of the most common examples is given by calcium silicate hydrolysis,
- 5 as described by following schematic reaction (Ebelmen, 1845; Urey, 1952):

6
$$CaSiO_3 + 2CO_2 + 3H_2O \rightarrow Ca^{2+} + H_4SiO_4 + 2HCO_3^-$$
 (2)

- 7 This equation represents the weathering of any silicate mineral into silicic acid (which often
- 8 precipitates as amorphous silica SiO₂), and consumes one more molecule of CO₂ than carbonate
- 9 dissolution while sending the same amounts of calcium and bicarbonate ions to the ocean. The
- 10 combination of equation (2) with calcium carbonate precipitation (the reverse of equation 1)
- shows how this results in a net removal of one molecule of CO₂:

$$CaSiO_3 + CO_2 \rightarrow CaCO_3 + SiO_2$$
 (3)

- 13 Weathering rates due to silicate hydrolysis tend to be considerably slower than from the
- dissolution of carbonate minerals it removes on average 0.28 to 0.30 Pg C per year (Amiotte
- 15 Suchet and Probst, 1995) hence the effect of atmospheric CO₂ consumption by silicate
- weathering only becomes a significant sink of carbon on geologic timescales (10⁵-10⁶+ years).
- 17 For the remainder of this article, dissolution of carbonates (on land) and hydrolysis of silicates
- will be treated separately and referred to as carbonate and silicate weathering, respectively.

1.3 Weathering in early carbon cycle models

- 20 Variations in rock weathering rates have long been believed to hold a major role in regulating the
- 21 Earth's long-term climate, and early (non-spatially explicit) carbon cycle models were built to
- 22 investigate the importance of the weathering feedback mechanism on various events in Earth's

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geological history. Walker, Hays and Kasting (1981), henceforth referred to as WHAK, 1 2 developed expressions relating silicate weathering rates to atmospheric pCO₂ (indirectly through vegetation productivity) and temperature (including a weak dependency on runoff) and used 3 them to offer a solution to the faint young Sun paradox by providing a convenient mechanism for 4 a slow and steady decrease in atmospheric greenhouse gas concentrations. Berner, Lasaga and 5 Garrels (1983), henceforth referred to as BLAG, also linked the rate of atmospheric CO₂ 6 7 consumption by silicate weathering to changes in surface air temperature, atmospheric partial CO₂ pressure, and river runoff and offered this dependency as a possible explanation for the 8 9 general decreasing trend of atmospheric CO₂ levels on geologic timescales. Although these 10 models used rudimentary parameterizations derived from early general circulation models and experimental data, they built a foundation for future long-term carbon cycle model studies. 11 12 Following BLAG, Berner (1991) built a geochemical cycle model in which the long-term 13 evolution of atmospheric carbon content would be driven by imbalances between CO₂ outgassing by volcanic activity and the burial of carbonate sediments following the weathering of silicate 14 rocks. The latter was given a dependency on air temperature and atmospheric CO2, and it was 15 used to solve a series of mass balance equations in order to determine the inward and outward 16 fluxes for the atmosphere-ocean, land, and mineralogical carbon reservoirs. This new model, 17 18 called GEOCARB, added more direct biological mechanisms (notably, the soil-biological enhancement of weathering) and introduced land elevation and runoff as independent 19 20 parameters. Much like its predecessor, this model was developed in order to reconstruct the 21 evolution of atmospheric pCO₂ over the past hundreds of millions of years. Subsequent versions were called GEOCARB II (Berner, 1994) and GEOCARB III (Berner and Kothavala, 2001), and 22 23 these further improved the weathering parameterizations based on the latest observational data

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1 and GCM output. They were later coupled with a model of atmospheric O₂ and ocean nutrients

2 to create COPSE (Bergman et al., 2004), a multi-element geochemical cycling model which

3 introduces a feedback-based interaction between biotic and abiotic elements of the Earth system.

1.4 Modelling the lifetime of anthropogenic CO₂

5 A number of studies have addressed the consequences of anthropogenic carbon emissions and

6 the multi-millennial lifetime of its perturbation on the climate system. The earliest attempt to

quantify the timescale of the weathering negative feedback mechanism can be traced back to

8 Sundquist (1991), who used a coupled atmosphere-ocean-carbon cycle model to obtain an e-

9 folding timescale on the order of several 10⁵ years. To this date, it remains the only study to

directly quantify this timescale, although values of 200 kyr (Archer et al., 1997) and 400 kyr

11 (Berner and Kothavala, 2001; Archer, 2005) have been cited in the literature. Most recent work

on this matter (Ridgwell and Hargreaves, 2007; Montenegro et al., 2007; Uchikawa and Zeebe,

13 2008; Archer et al., 2009; Eby et al., 2009) has involved intermediate-complexity models, which

are usually limited in scope to 10⁴ years or shorter. Hence the weathering feedback mechanism

in these studies is limited to the pH neutralization effect of carbonate weathering on the oceans

16 (which restores the lysocline to its original depth), and silicate weathering is either ignored

17 altogether or prescribed as a global constant average flux.

In a pioneering study, Walker and Kasting (1992) considered the impact of the rock cycle and

carbonate sediment dissolution in projections of future changes in the global carbon cycle. Their

model was built on the assumption that the dependency of carbonate and silicate weathering rates

to changes in the carbon cycle (aka. atmospheric CO2 levels) was purely abiotic, which was in

22 line with the other geochemical cycling models of the time. Following on Walker and Kasting

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1 (1992) as well as the recent innovations in COPSE, Lenton and Britton (2006) posited that

2 biological changes in the Earth system could further enhance the increase or decrease in rock

weathering rates, especially in the context of a rapidly warming world which would likely result

from unabated anthropogenic emissions. Their carbon cycle model included sophisticated biotic

and abiotic transports of carbon, introducing a box-model representation of carbonate and silicate

weathering processes in which weathering rates were directly dependent on plant productivity,

rather than on atmospheric CO₂ concentrations. This allowed them to investigate the role of land

8 use changes on the long-term recovery of atmospheric CO2; in particular, they found that

vegetation-suppressing land use changes would force CO₂ levels to stabilize above preindustrial

levels on geologic timescales, thus indefinitely trapping some of the anthropogenic emissions in

11 the atmosphere.

12 1.5 On spatially-explicit weathering models

13 Few attempts have been made to explore the spatial variability of carbonate and silicate

weathering rates and how it may affect the global efficacy of the weathering negative feedback

mechanism. The main problematic relating to the development of spatially explicit weathering

schemes is the necessity to compute weathering individually for each land grid cell, which is

entirely more complex than using a globally-averaged value, for which many precedents exist in

the literature. The GEM-CO₂ model (Amiotte Suchet and Probst, 1995) addressed this problem

by defining the spatial variability in terms of rock types, and using data for bicarbonate (HCO₃⁻)

20 concentration and runoff collected over various mono-lithologic drainage basins (Meybeck,

1987) to establish empirical linear relationships between weathering flux and runoff for a series

of major rock types. Arguing that these two factors (runoff and rock type) were the main factors

controlling the consumption of atmospheric/soil CO₂ by weathering, they calculated the global

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distribution of CO₂ consumption. Their results showed a higher intensity of weathering in the 1 2 Northern Hemisphere (due to rock type) and in equatorial regions (due to runoff). They later refined the global distribution of rock types by attributing one of six rock types to each land unit 3 4 of a 1°×1° grid (Amiotte Suchet et al., 2003); this distribution will be used as the basis for our 5 spatially explicit weathering scheme. Rock types, from having the smallest to largest impact on weathering, are classified as follows: plutonic and metamorphic (shield) rocks, sands and 6 sandstones, extrusive igneous (acid volcanic) rocks, basalts, shales and evaporites, and carbonate 7 rocks. The latter designs a loose group of predominantly carbonate-based rocks, and of the other 8 9 five rock types only sandstones and shales contain a fraction of carbonate-weathered minerals. 10 In the other rock types, the prevalence of carbonate minerals is too variable and difficult to estimate, hence they are assumed to contain only silicate-weathered minerals. Using the GEM-11 CO₂ model, carbonate rocks and shales were found to both consume 40% of the total continental 12 CO₂ uptake despite occupying a much smaller fraction of land area, while sandstones and shield 13 rocks contributed much lower than their outcrop abundance. A similar rock type distribution 14 was developed (Gibbs and Kump, 1994; Bluth and Kump, 1994) (hence GKWM), using both an 15 16 empirical linear coefficient and an exponential factor to express weathering dependence on runoff for different rock types; however the results produced by their lithological distribution 17 was found to be very similar to that of GEM-CO₂, in terms of global weathering intensity and the 18 19 consumption of atmospheric/soil CO₂ (Colbourn et al., 2013). 20 Other instances of spatially-explicit weathering models are few in the literature. The GEOCLIM 21 model has a built-in two-dimensional weathering scheme that has been used to investigate the climatic impacts of tectonic continental reorganization and weathering-vegetation interactions 22 (Donnadieu et al., 2009). More recently, a spatially-explicit scheme was added to the GENIE 23

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1 model (Colbourn et al., 2013), using lithological databases from the GEM-CO₂ and GKWM

2 models, temperature dependency from the GEOCARB models, NPP dependency as introduced

3 by Lenton and Britton (2006), and runoff dependency from GEM-CO₂. Although the paper

4 focused mostly on exploring the various model options, the authors were able to simulate the

5 entirety of the climate system recovery from a 5000 Pg C anthropogenic pulse at year 2000,

6 showing that within 0.5-1 Myr the atmospheric CO₂ levels would return to pre-industrial levels.

7 2 Methods

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8 2.1 Climate model description

9 In this study we used version 2.9 of the University of Victoria Earth System Climate Model

10 (henceforth UVic ESCM, or UVic model), which is an intermediate complexity coupled

11 atmosphere/ocean/sea-ice model with integrated land surface and vegetation schemes (Weaver et

al., 2001). Its main component is version 2.2 of the GFDL Modular Ocean Model (MOM), a

three-dimensional ocean general circulation model with 19 uneven vertical levels (Pacanowski,

1995), which is coupled to a vertically integrated energy-moisture balance atmosphere model

(Fanning and Weaver, 1996), a dynamic-thermodynamic sea-ice model (Bitz et al., 2001), a land

surface scheme and dynamic global vegetation model (Meissner et al., 2003), and a

sedimentation model (Archer, 1996). Land surface properties (surface temperature, soil moisture

content and temperature, and snow cover) and soil carbon content are computed with a single (1-

19 meter) layer version of the Meteorological Office Surface Exchange Scheme version 2 (MOSES-

20 2) (Cox et al., 1999), and terrestrial vegetation dynamics are handled by the Hadley Centre's

Top-down Representation of Interactive Foliage and Flora Including Dynamics (TRIFFID)

model (Cox, 2001). TRIFFID describes the state of the terrestrial biosphere in terms of soil

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1 carbon content and vegetation distribution, which is expressed through the structure and

2 coverage of five plant functional types: broadleaf tree, needleleaf tree, C₃ grass, C₄ grass, and

3 shrub vegetation.

4 The UVic ESCM also includes a fully coupled global carbon cycle, which consists of inorganic

5 carbon chemistry and land-surface exchanges of CO₂ (Ewen et al., 2004), and a Nutrient-

6 Phytoplankton-Zooplankton-Detritus (NPZD) module which calculates the contribution of the

7 biological pump to ocean biogeochemistry (Schartau and Oshlies, 2003; Schmittner et al., 2008).

8 Terrestrial carbon fluxes and reservoirs are described by Matthews et al. (2005), and coupled to

9 the global model by Meissner et al. (2003).

10 The model is driven in the short term by seasonal variations in solar insolation and wind fields

(Kalnay et al., 1996), and in the long-term by orbital parameter changes and a reconstruction of

atmospheric CO₂ content over the past 20 thousand years (Indermühle et al., 1999). The spatial

coverage and height of continental ice sheets is prescribed every 1000 years using data from the

model ICE-5G (Peltier, 2004); thus these ice sheet configurations also serve to drive climate

15 changes during glacial periods. The land-sea configuration used in all sub-components operates

in a global spatial domain with a spherical grid resolution of 3.6° (zonal) by 1.8° (meridional),

which is comparable to most coupled coarse-resolution AOGCMs.

2.2 Weathering model description

19 Terrestrial weathering in the UVic model is parameterized as a land-to-ocean flux of dissolved

inorganic carbon (F_{DIC}) and alkalinity (F_{ALK} , with $F_{ALK} = 2F_{DIC}$) via river discharge. In the

21 standard version of the model, the incoming flux of carbon to the ocean as weathering is set to

22 equal the sedimentation rate of CaCO₃ in order to balance the long-term carbon and alkalinity

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budgets in the ocean; the initial, steady-state value is typically held constant throughout the

2 transient model runs. This effectively suppresses the long-term negative feedback mechanism by

preventing the weathering rate from adapting to changes in environmental factors such as

4 temperature and atmospheric CO₂ concentration. Meissner et al. (2012) replaced the standard

5 parameterization of weathering in the UVic model with a number of adaptations from previous

carbon-cycle box models in order to investigate the role of rock weathering as a carbon sink for

anthropogenic carbon emissions. They found that the long-term climate response to various

8 emission scenarios depends almost exclusively on the total amount of CO₂ released regardless of

9 the rate at which it is being emitted, and carbon uptake through an increase in terrestrial

weathering has a significant effect on the climate system. There were, however, some

differences between the various weathering schemes concerning the rate of carbon removal.

12 In this section we describe a spatially explicit weathering scheme developed for use within the

UVic ESCM. Steady-state carbonate and silicate weathering rates are calculated for each land

grid cell based on the local rock type (Sect. 2.2.1) and runoff (Sect. 2.2.2). In transient model

runs, these values are modulated by changes in temperature, atmospheric CO2 concentrations or

vegetation productivity, and runoff (Sect. 2.2.3), which are updated on each time step based on

model output. Changes in carbonate and silicate weathering rates are returned to the model in

the form of a riverine flux of carbon and alkalinity (Sect. 2.2.4), which is routed to the ocean.

19 *2.2.1 Worldwide distribution of rock types*

20 The two-dimensionality of the weathering model is rooted in the uneven distribution of rock

types across the world. Thus, regions with more active lithologies (mostly sedimentary rocks

22 such as carbonates and shales) yield higher weathering rates under similar climate conditions,

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and these are more sensitive to changes in climate controls than regions predominantly covered 1 2 by weathering-resistant lithologies (igneous and metamorphic rocks, basalts and granites). Whereas the worldwide distribution of continental rock lithology is well known, there is only 3 limited knowledge of the impact of different rock types on the amounts of riverine exports, 4 therefore any estimation of weathering rates based on local lithological composition is subject to 5 some discrepancy. Two of the most prominent spatially explicit weathering schemes, the Gibbs 6 7 and Kump weathering model (Gibbs and Kump, 1994), and GEM-CO2 (Amiotte Suchet and Probst, 1995), each use their own set of land lithological data, which classify the entirety of the 8 9 world's lithologies into one of several rock types, defining the impact of each on weathering by 10 modifying its basic dependency on runoff. While developing a 2-D weathering model for use into the climate model GENIE, Colbourn et al. (2013) compared the output generated by both 11 lithological distributions. They found that the end result did not differ much between the two 12 models (they found a difference of only 4 ppm in atmospheric CO₂ concentration 100 kyrs 13 following a pulse of 5000 Pg C in the atmosphere), concluding that differences between 14 individual rock distribution datasets have a negligible impact on the model output. 15 In this study we used the lithological distribution paradigm first introduced in GEM-CO2, and 16 later published by Amiotte Suchet et al. (2003). The flux of atmospheric/soil CO2 from 17 18 chemical weathering on each continental grid cell was given an empirical linear relationship to runoff (see Sec. 2.2.2) depending on its assigned predominant rock type, which was classified as 19 one of six different lithological categories: sands and sandstones, shales, carbonate rocks, shield 20 21 rocks, acid volcanic rocks, and basalts. Sedimentary rocks (limestones, shales, sandstones) contain significant amounts of carbonate rocks, and thus do not consume atmospheric CO2 as 22 efficiently as other rock types, despite sending a higher riverine flux of weathering products. 23

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- 1 The adaptation of the rock type distribution map to the UVic model is shown in Figure 1. The
- 2 spatial resolution of the UVic model (3.6°×1.8°) is about 6.5 times coarser than that of the
- 3 original database (1°×1°) hence the adapted rock distribution paradigm was defined according to
- 4 the partitioning of rock types within the area contained by each UVic model grid cell. The
- 5 resulting runoff multiplier and carbonate to silicate fractionation therefore becomes a weighted
- 6 spatial average of all of rock type multipliers in Table 1.
- 7 2.2.2 Calculating the steady-state weathering rate
- 8 The reference weathering rate is calculated for each individual grid cell based on local steady-
- 9 state runoff R_0 (Figure 2a) and rock type composition. Following Amiotte Suchet and Probst
- 10 (1995), the local riverine fluxes of bicarbonate ions from carbonate (f_{Ca}) and silicate (f_{Si})
- 11 weathering are computed as:

$$f_{Ca} = R_0 \sum_{i} frac_i k_i \alpha_i \tag{5}$$

$$f_{Si} = R_0 \sum_{i} frac_i k_i (1 - \alpha_i)$$
 (6)

- where $frac_i$ is the fraction of rock type i present in the grid cell, k_i is the rock type specific
- weathering rate multiplier, and α_i is the fraction of rock type given to weather as carbonate
- 14 rocks. The different rock types and their weathering parameters are shown in Table 1. The
- weathering rate multipliers (k_i) were derived from the data by Amiotte Suchet et al. (2003) and
- the fractionation of rock types between carbonate and silicate rocks is adapted from the work of
- Gibbs et al. (1999), following the interpretation of Colbourn et al. (2013).
- 18 The resulting steady-state carbonate and silicate weathering rates at pre-industrial (1800AD)
- 19 conditions are shown in Figure 2b. There is a noticeable concentration of CaCO₃ weathering in

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1 areas of high runoff with bedrock composed predominantly of carbonate rocks (for example,

2 Southeast Asia), whereas CaSiO₃ weathering is spread more evenly across the world. It is

3 noteworthy that the Amazon basin features by far the highest runoff yet produces unremarkable

weathering rates (compared to other tropical areas) due to the prevalence of the weathering-

5 resistant shield rocks. The same observation can be used to explain the low weathering rates in

central Africa. This weathering distribution compares reasonably well with the CO₂

consumption distribution found by Amiotte Suchet and Probst (1995), but it doesn't reproduce

8 the large values at northern high latitudes (especially in northern Asia) that can be found using

9 the GEM-CO₂ model. The distribution of bicarbonate fluxes of Gibbs and Kump (1994) displays

a somewhat lower equator-to-pole gradient in weathering rates, and suggests an area of high

weathering in the southeast USA which is not reproduced with our model, mainly on account of

low runoff in the region. These discrepancies are likely due to precipitation bias in the UVic

13 ESCM. However, both models appear to agree with our finding that southeastern Asia is the

region with the highest regional weathering intensity. Globally, the 2-D weathering scheme

sends a DIC flux of 0.166 Pg C/y into the ocean, which is approximately 15% more than the 0-D

model output (0.145 Pg C/yr) (Meissner et al. 2012), and on par with previous estimations of

pre-industrial global weathering intensity.

18 2.2.3 Modulation of weathering rate

19 In transient model simulations, the carbonate and silicate weathering rate for each grid cell is

20 modulated by changes in local environmental conditions. They were made dependent on surface

21 air temperature, atmospheric carbon dioxide content, and runoff in a similar manner to previous

carbon cycling models. Following Lenton and Britton (2006), we have included the option of

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- 1 replacing the dependency on CO2 concentration by vegetation productivity, which more directly
- 2 accounts for the impact of biological factors on weathering intensity.
- 3 Temperature is a known controller of weathering intensity as higher temperatures increase the
- 4 kinetic energy of molecules, facilitating the atomic encounters which lead to the chemical
- 5 dissociation of minerals. Although it is impossible to derive a relationship between temperature
- 6 and weathering rates from first principles, laboratory and field studies have correlated the
- 7 concentration of bicarbonate ions in a solution to water temperature in order to develop an
- 8 empirical formulation. For carbonate weathering, we used the results of Harmon et al. (1975),
- 9 who compared the groundwater temperature and bicarbonate ion concentration of several North
- 10 American watershed to come up with the following empirical relationship:

$$g_{cq}(SAT) = 1 + 0.049(SAT - SAT_0) \tag{7}$$

- where SAT and SAT_0 are the transient and steady-state surface air temperature, respectively. For
- 12 silicate weathering, we used a version of the Arrhenius rate law of Brady (1991) which was
- adapted into the RokGeM by Colbourn et al. (2013):

$$g_{Si}(SAT) = e^{0.09(SAT - SAT_0)}$$
 (8)

- Here, the constant of 0.09 inside the exponential expression was obtained using an activation
- energy of 63 kJ mol⁻¹ for silicate weathering and a global initial temperature of 288K (global
- 16 average pre-industrial temperature). The activation energy is poorly constrained, but has been
- shown to have little effect on the long-term consumption of atmospheric CO₂.
- 18 The productivity dependence of weathering serves to illustrate the biological and soil-
- 19 enhancement factors which control weathering intensity, with vegetation net primary
- 20 productivity a suitable proxy for biological activity in an area. Lenton and Britton (2006)

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- 1 introduced a simple linear dependence of weathering on productivity based on the steady-state
- 2 proportion of global productivity to global weathering flux:

$$g(NPP) = \frac{NPP}{NPP_0} \tag{9}$$

3 where NPP and NPP₀ are the transient and steady state net primary production, respectively,

4 taken explicitly from the output of the coupled land surface scheme MOSES-2 and vegetation

5 module TRIFFID. This formulation works reasonably well in 0-D models with globally-

6 summed values of productivity and weathering rate; however, some problems arise when trying

7 to use it in a spatially-explicit model due to its inherent assumption that productivity and

8 weathering intensity are directly related at steady state. A good example of this would be at the

9 continental margins of predominantly ice-covered continental landmasses (Greenland,

Antarctica) where some of the land may be ice-free, but too cold to support any vegetation.

However, the presence of nearby ice sheets generates a meltwater runoff flux which greatly

12 enhances weathering, in spite of the lack of vegetation. Therefore, any expansion of vegetation

in this area, however small (which is not unrealistic given the extreme warming scenarios

14 examined here), would result in an unreasonable increase in weathering. In order to rectify this

situation, we have introduced a modified version of equation 9 which calculates the increase in

local weathering rate when *NPP* is greater than its steady-state value:

$$g(NPP) = \begin{cases} NPP/NPP_0, & NPP < NPP_0 \\ (NPP - NPP_0) / NPP_{0,global} \\ 1 + \frac{f_0}{f_{0,global}}, & NPP \ge NPP_0 \end{cases}$$
(10)

where $f_0 = f_{Ca} + f_{Si}$ and the "global" indices indicate the globally-summed value of that

18 variable. The right-hand term in the brackets is a compensation term, which modifies the

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1 increase in weathering based on the relative contribution of the grid cell to the global

2 productivity compared with its contribution to the global weathering flux. This results in a

3 redistribution of NPP-induced changes in weathering without changing the globally-summed

increase in weathering intensity (this is true only in the absence of other controlling factors, such

5 as temperature). Note that the parameterization is unchanged from equation (9) whenever NPP

6 is lower than its initial value, mostly to avoid computing negative values of g(NPP). This has a

7 relatively benign impact on the global result, as the values calculated from equation (10) only

8 differ significantly from those of equation (9) when NPP is much greater than NPP₀.

9 As an alternative to productivity dependence, we also included the option to parameterize

10 weathering as a function of atmospheric CO₂ content, following the approach used in the

11 GEOCARB II model of Berner (1994):

$$g(CO_2) = \left(\frac{2\frac{pCO_2}{pCO_{2,0}}}{1 + \frac{pCO_2}{pCO_{2,0}}}\right)^{0.4}$$
(11)

where pCO_2 and $pCO_{2,0}$ are the transient and steady-state atmospheric concentration of CO₂,

13 respectively. This relationship has long been used to estimate the fertilizing effect of CO₂ on

14 land plants, and thus becomes here an indirect parameterization of the biological enhancement of

weathering. It can be used *in lieu* of equations 9 and 10 as a model option.

Runoff is the most widely used factor of weathering intensity as it constitutes a good proxy for

17 the strength of the water cycle in an area. One may consider the fact that high runoff

environments tend to be associated with intense weather activity (the rainforests, for example),

19 and also that stagnant waters quickly become saturated, thus limiting the efficiency of

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- 1 weathering. Given that we already include runoff in the calculation of steady-state weathering,
- the transient runoff dependency is a simple adjustment following *Berner* [1994]:

$$g_{Ca}(R) = \frac{R}{R_0} \tag{12}$$

$$g_{Si}(R) = \left(\frac{R}{R_0}\right)^{0.65} \tag{13}$$

- 3 where R and R_0 are the transient and steady-state river runoff, respectively, which are also taken
- 4 explicitly from the output of the land surface scheme MOSES-2. The difference between the
- 5 formulations for carbonate and silicate weathering is an empirical correction based on the
- 6 assumption that bicarbonates from the weathering of silicate rocks are more diluted in rivers than
- 7 for carbonate weathering. The value of 0.65 in equation (13) was taken from Berner (1994);
- 8 although the value itself has a large margin of error, it has been shown to have only a modest
- 9 effect on the overall efficiency of the weathering feedback mechanism.
- 10 To summarize, we have developed a two-dimensional weathering scheme whereby the steady-
- state values of carbonate and silicate weathering fluxes (see Sect. 2.2.2) are modulated by
- 12 changes in temperature, vegetation productivity (alternatively: atmospheric CO2 concentration),
- and runoff. Thus the complete weathering parameterizations take the form:

$$F_{Ca} = f_{Ca} \cdot g_{Ca}(SAT) \cdot g(NPP) \cdot g_{Ca}(R) \tag{14}$$

$$F_{Si} = f_{Si} \cdot g_{Si}(SAT) \cdot g(NPP) \cdot g_{Si}(R) \tag{15}$$

- Weathering is calculated in each individual land grid cell, and routed to the coastal ocean as
- 15 fluxes of alkalinity and dissolved inorganic carbon, explained in section 2.2.4.
- 16 2.2.4 Effects of weathering on ocean biogeochemistry

(which is not included in the UVic model):

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In the UVic ESCM, weathering does not have a direct impact on atmospheric or land surface carbon; its effects are prescribed through the riverine exports of weathering products which are sent to the ocean and modify its chemical composition. The chemical weathering processes are described by equations 1 (carbonate) and 2 (silicate), both resulting in a flux of two moles of bicarbonate ions (HCO_3^-). The flux of dissolved inorganic carbon (F_{DIC}) is counterbalanced by the consumption of atmospheric carbon dioxide during the weathering reactions, leaving a net DIC flux of one mole for carbonate weathering, and none for silicate weathering; a constant term

was also added to represent the contribution of volcanic outgassing to global carbon emissions

$$F_{DIC} = F_{Ca} + F_{volc} \tag{16}$$

10 Given that, in the absence of external forcings, the CO₂ consumption by silicate weathering is 11 meant to counter the intake of carbon from the geologic reservoir from volcanic eruptions, we set the constant term F_{volc} to equal the steady-state flux of silicate weathering $(F_{volc} = F_{Si,0})$. 12 13 Meanwhile, the net flux of alkalinity (F_{ALK}) remains equal to the flux of bicarbonate ions. The above discussion is summarized in the following set of equations, which describes the 14 15 partitioning of carbonate weathering and silicate weathering fluxes into dissolved inorganic carbon and alkalinity fluxes, which are then globally summed and fed to the ocean 16 biogeochemistry module: 17

$$F_{DIC} = F_{Ca} + F_{Si,0} \tag{17}$$

$$F_{ALK} = 2F_{Ca} + 2F_{Si} \tag{18}$$

Note that our choice of F_{volc} effectively equilibrates ocean biogeochemistry during equilibrium runs ($F_{ALK,0} = 2F_{DIC,0}$). The values calculated here represent net fluxes over the entire surficial (atmosphere-land-ocean) reservoir, and in particular the simplification for the net flux of DIC is

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1 based on the assumption that the consumption of CO₂ from the atmosphere is immediately

2 balanced by an equivalent uptake of carbon from the ocean. This would be true in general, given

3 that the timescale of the weathering negative feedback mechanism far exceeds that of

4 atmosphere-ocean mixing; however in the timescales considered here (10³-10⁴ yrs) there would

5 be some delay between the consumption of CO₂ from the atmosphere and the release of CO₂ in

6 the ocean following CaCO₃ burial. This delay would not significantly alter the impact of

7 weathering on atmospheric geochemistry, but could reduce by as much as 10% the rate at which

8 alkalinity increases in the ocean (Colbourn et al., 2013).

9 2.3 Steady-state weathering and description of transient model simulations

10 Pre-industrial steady-state weathering was obtained by integrating the model for over 20,000

years under year 1800 boundary conditions, using rock type dependency and distribution as

detailed in sections 3.1.1 and 3.1.2. Land-to-ocean weathering fluxes stabilized in less than 10³

13 years, on account of runoff being mostly computed from atmospheric output. However, the

14 fixing of deep ocean alkalinity and dissolved inorganic carbon (DIC) content would have

required as much as 10^5 model years – an impossibly long simulation time given the level of

complexity of the UVic model. Hence we extracted the model steady state after 10⁴ years, but

kept the background steady-state run ongoing concomitantly with the transient model

simulations in order to correct the output of the latter based on changes in the former. Each

transient simulation was forced with the historical natural and anthropogenic carbon emission for

20 years; at year 2000 an additional 5000 Pg C were emitted over one year (unless otherwise

indicated), and carbon emissions were set to zero thereafter. All simulations were carried out for

a period of ten millennia, ending at year 12,000.

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1 A total of eight model versions were integrated to year 12,000, which we classified into three

2 groups of experiments (see Table 1 for a description of all experiments). Group A (Section 3.1)

3 experiments investigate the impact of the intensity and span of the prescribed carbon emissions.

4 Simulation A0 is the basic emission scenario outlined in the above paragraph, and thus served as

5 the main control run for this paper. Simulation A1 is similar but used a more conservative

estimate of 1000 Pg C for future anthropogenic emissions. Finally, simulation A2 extended the

carbon emission total of 5000 Pg over a much longer period: emissions were increased linearly

8 until reaching double the current (year 2000) carbon emissions; the remaining carbon emissions

were then distributed evenly during the period from 2050 to 3000, then set to zero thereafter.

10 Although distributed over a longer period, the total carbon emissions remained unchanged from

our control run A0.

12 Group B experiments (Sect. 3.2) compare the various model representations of the biological

enhancement factor. In simulation B1, we replaced the NPP dependence term g(NPP) in

equations 14 and 15 with $g(CO_2)$ from equation 11 on all grid cells. Although carbon dioxide

concentrations in the atmosphere are known to vary slightly across the surface of the Earth, in

the UVic model pCO_2 is a global term with no defined spatial variability. This effectively

removes the two-dimensionality of the biological feedback term, leaving temperature as the sole

spatially-explicit variable. Runoff does not vary much unless there are major changes in

hydrology or ice sheet distribution, neither of which were considered in our simulations. A

further simplification was made in simulation B2 by removing the biological enhancement factor

altogether and incorporating a parameterization that is only based on the temperature-dependent

22 part of our spatially-explicit scheme.

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1 Group C experiments (Sect. 4.3) compare the relative importance of carbonate and silicate

2 weathering through their impacts on riverine fluxes of alkalinity and DIC. In simulation C1, we

eliminated the silicate weathering feedback ($F_{Si} = f_{Si}$), leaving only the carbonate weathering

4 part of the parameterization. Likewise, in simulation C2, the carbonate weathering feedback was

negated ($F_{Ca} = f_{Ca}$), isolating the impact of the silicate weathering feedback. Finally, in model

6 version C3 we eliminated both carbonate and silicate weathering feedbacks to maintain constant

weathering fluxes (at steady-state values) throughout the simulation. This last model version

effectively simulates the carbon sequestration potential of the oceans in the absence of the

9 weathering feedback mechanism.

10 For each of the model versions outlined above (with the exception of C1 and C2), an identical

11 setup was used with a zero-dimensional version of the weathering model whereby weathering

12 rates were calculated based on global, rather than local changes in the control parameters

(temperature, NPP, runoff); these 0-D model versions are identified in the figures using the "*"

notation (for example, "A0*" refers to the zero-dimensional version of simulation A0). The code

for these 0-D model versions was developed in an earlier study of terrestrial weathering changes

with the UVic model (Meissner et al., 2012).

3 Results

3.1 Group A results

19 The time series of CO₂ concentration in the atmosphere as well as weathering fluxes of carbon

and alkalinity are shown in Figure 3a for each of the pulse (group A) scenarios (solid lines), and

21 compared with results from similar scenarios using the 0-D version of the model (dotted lines).

22 For all simulations, the 2-D model was always more efficient in removing CO₂ from the

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atmosphere than its 0-D counterpart. This can be partially explained by the initial global total 1 2 weathering being slightly higher in the 2-D model (see Section 2.2.2); however, this cannot account for alkalinity weathering increasing nearly three times as much in the 2-D model as it 3 does in the 0-D version (interestingly, 0-D weathering rates seem to be slightly higher than 2-D 4 values for scenario A1). Instead, we propose that this is a natural consequence of using a two-5 dimensional approach. Temperature, productivity, and runoff are closely related, as all three are 6 7 positively affected by the increase in atmospheric CO₂: temperature from the greenhouse effect, 8 NPP through the CO₂ fertilization of plants, and runoff as a result of both a temperature (hence 9 precipitation) increase and the CO₂-induced increase in plant water-use efficiency. Vegetation 10 productivity also reacts positively to increases in temperature in extratropical regions, although this effect could be overcompensated for by an opposite reaction in tropical regions, where 11 temperatures exceed the threshold for optimized plant growth (Matthews et al., 2005; Matthews 12 et al., 2007). This means that areas which see a large increase in one variable will more often 13 than not see equally large increases in one or both of the other variables, further enhancing the 14 local increase in weathering rates. A zero-dimensional model would not be able to create this 15 16 effect because it uses globally summed or averaged variables. This is especially important with regards to temperature, as the global average would be dominated by oceanic SAT changes, 17 18 which tend to be smaller than continental SAT changes. Scenario A2, when compared to A0, suggests that the ability of the Earth system to recover from 19 20 anthropogenic emissions is essentially independent of the rate at which the emissions occur. 21 Atmospheric CO₂ concentrations recovered more slowly in the gradual emissions scenario at first since weathering fluxes were not increased as much, but the gap between the two curves 22 gradually narrowed after A2 emissions ended at year 3000. We found a difference of 164 ppm in 23

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in which a 5000 Pg C pulse was spread over 300 years. Given that the longer the carbon 3 emissions are spread out over time, the longer it takes for atmospheric CO₂ levels to catch up to a 4 pulse scenario, we can surmise that pulse scenarios would overestimate the ability of the 5 6 weathering feedback mechanism to remove carbon from the atmosphere in the next several 7 millennia, unless a way is found to mitigate anthropogenic emissions within the next century. 8 Time series of various carbon reservoirs for Group A scenarios are shown in Figure 3b. The 9 ocean reservoir content at year 12,000 was nearly identical for scenarios A0 and A2, indicating that the ocean was even more indifferent to the rate of carbon emissions than the atmosphere. 10 Zero-dimensional model ocean carbon exceeded the 2-D output at around year 8500 for 11 12 scenarios A0 and A2, due to the fact that there was more carbon remaining in the atmosphere-13 ocean system. It is interesting that the land and sediment carbon reservoirs behaved differently 14 from other reservoirs. The large carbon emissions (and associated temperature anomalies) in A0 15 and A2 appeared to have a counterproductive effect on land carbon content, which was not seen

under the more modest temperature increase of simulation A1. If anything, this results points out

the inability of the land reservoir to store any significant amount of excess carbon from the

atmosphere on millennial timescales. Finally, the sediment carbon curve also behaved somewhat

counterintuitively, as all three 2-D pulse scenarios produced a comparable increase of sediment

carbon content, regardless of emission rate or amount of carbon released. For the 0-D model

versions, total CaCO₃ buried mass increased more rapidly for scenario A1 despite lower amounts

of carbon emitted. These results arise because carbon burial depends on a delicate balance which

involves ocean temperature, alkalinity, and calcite concentration. Oceans in scenario A1

atmospheric CO₂ concentration between A0 and A2 at year 12,000, which is nonetheless much

greater than the difference between the pulse and IPCC A2 simulations in Meissner et al. (2012),

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1 contained less CO₂ and CaCO₃, but were also cooler than in A0 and A2, which may explain why

2 the total accumulation of CaCO₃ sediments was comparable between all three scenarios.

3 Global changes in surface air temperature, vegetation NPP, and surface runoff are shown in

4 Figure 4 at various times during the 10,000 year simulation. As seen in Figure 4a, the most

5 significant changes in temperature mostly occurred poleward of 60 degrees of latitude; however,

6 there were also increases in many tropical regions. These results are to be expected given the

static nature of wind fields in the UVic model, which prevent a reorganization of atmospheric

circulation and thus trap the warm anomalies in the tropics. Figure 4a (see also Figure 3c) also

9 reveals that the cooling effects of carbon sequestration were not felt until well after year 3000,

despite atmospheric CO₂ concentrations being decreased by nearly 1000 ppm between years

11 2000 and 3000; this is simply due to the thermal inertia of the ocean (Matthews and Caldeira,

2008). By year 12,000, temperature anomalies across the globe became fairly uniform, with

every area averaging 1-2°C warmer than pre-industrial state.

14 Changes in vegetation net primary productivity are shown in Figure 4b. Most of the world saw

an increase in vegetation activity from the direct effect of CO₂ fertilization, with the exception of

desert areas which remained the same (Africa, Asia) or become more arid (Australia). Changes

in NPP also correlated well with changes in river runoff (Figure 4c); this is mainly a

consequence of the effect of increased CO₂ concentrations on plants, which optimises vegetation

water-use efficiency, leading to an increase in soil moisture and therefore runoff (Nugent and

20 Matthews, 2012; Cao et al., 2010). The very large NPP increase in Indonesia around year 3000

21 was likely caused by the replacement of rainforest by the much more productive C4 grasses, and

22 further enhanced by a 1000-year legacy of high CO₂ fertilization.

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1 The resulting impacts of these changes in temperature, vegetation productivity, and runoff on

2 CaCO₃ and CaSiO₃ weathering are shown in Figure 5. We found that changes in weathering

were more strongly correlated to changes in NPP, and to a lesser extent, runoff. The influence of

rock type distribution was also noticeable, especially on the carbonate/silicate weathering

partitioning, while temperature had an overall modest impact. Most areas saw a moderate to

high increase in both weathering types, with the exception of arid areas (deserts and ice caps)

which experienced a minor decrease in weathering. The most significant change occurred in

8 central western Asia (Kazakhstan), mirroring a moderate increase in vegetation productivity

coupled with a considerable temperature change during the third millennium CE. The anomaly

all but disappeared in later snapshots of the simulation, once global temperatures were no longer

warm enough to sustain such high levels of vegetation productivity. Indonesia also saw a large

increase in silicate weathering rates, on account of all three controlling parameters increasing by

a large margin in the area, coupled with a predominantly silicate-heavy lithology. In later stages

of the model simulation, weathering rate anomalies had mostly retreated to the tropical latitudes,

where productivity and runoff anomalies persisted the longest; elsewhere the increase in

weathering rates was reduced to below 10% of their value during the third millennium CE.

3.2 Group B results

18 The purpose of this group of experiments was to assess the importance of including a

parameterization for NPP (A0) rather than atmospheric CO₂ concentration (B1), or a weathering

20 scheme based exclusively on temperature and runoff feedbacks (B2). Model output for

atmospheric CO₂ concentration and weathering fluxes is shown in Figure 6a. The results

strongly suggest that using vegetation productivity rather than CO₂ as a proxy for biological

activity makes weathering fluxes much more sensitive to overall climate and environmental

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changes. Weathering fluxes peaked around year 2200, and the increase for simulation A0 (using 1 2 NPP) was twice that for simulation B1 (using CO₂), and about three times larger than for simulation B2 (with no biological effect); in other words, adding a NPP dependence tripled the 3 weathering increase compared to the case using a temperature dependence only. This is likely a 4 result of rapid vegetation expansion in the high latitudes and the appearance of warm-adapted 5 and more productive biomes in the temperate regions, which was taken into account in A0 but 6 7 not in the Group B model versions. As a result, the recovery time of atmospheric CO₂ levels was much faster in A0, and vegetation productivity rapidly dropped below the levels of B1 and B2 8 (not shown). Interestingly, after year 7000 the weathering DIC flux in simulation A0 fell below 9 10 that of B1, indicating that from that point onward the parameterization in B1 was more effective in removing CO₂ from the atmosphere. This feature does not appear in the 0-D model results, 11 where DIC weathering fluxes always remained higher in the productivity-dependent model 12 version. As mentioned in section 3.1, it is possible that the increased effectiveness of the 2-D 13 weathering parameterization (compared to 0-D) is caused by the coincidence of large 14 temperature/runoff increases within areas that also see a large increase in vegetation NPP. 15 Figure 6b displays the time series of ocean and sediment carbon, as well as three parameters 16 which are used to analyze the evolution of calcite sedimentation in the model: CaCO₃ 17 18 production, pore layer portion, and dissolution. In contrast to 0-D versions of the model, there was a clear convergence of all three 2-D curves for ocean carbon content resulting from a 19 20 substantial drop in weathering rates during the latter half of the simulation period. The rate of 21 increase of CaCO₃ buried mass was slower in model versions with lower weathering rates; this surprisingly differs from Fig 3b where there was not much difference between the three model 22 versions. However, the 0-D curves still displayed a significant lag behind their 2-D counterparts. 23

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1 The pore layer portion remained unchanged during the first thousand model years after the

2 introduction of the 5000 Pg C pulse, thus mirroring the results of *Meissner et al.* [2012]. Higher

biological activity and carbonate concentration in the surface ocean due to warmer temperatures

4 was found to lead to a sharp increase in calcium carbonate formation and precipitation; this was

balanced by a rising of the carbon compensation depth (CCD) in the deep ocean fueled by the

rising acidity of the ocean, which increased the overall dissolution rate of calcite. As the more

7 immediate effects of the carbon emission pulse receded, oceans became cooler and calcite

8 formation weakened, while dissolution kept increasing for another 1000 years. This created an

unbalance in the CaCO₃ pore layer fraction which appears from year 4000 onward. Note that

even though dissolution rates in the deep ocean exceeded calcite production in the surface layer,

there was still an overall increase in CaCO₃ buried mass due to the enormous increase in oceanic

carbon content.

13 Figure 7 displays the spatial distribution of CaCO₃ and CaSiO₃ weathering changes at various

points of the simulation timeline for model version B1. Several areas of higher weathering from

15 Figure 5 were completely absent (central Eurasia), and some others were greatly reduced

(tropical Africa). These are the most important examples of how the vegetation productivity

parameterization can greatly enhance carbonate weathering locally, and silicate weathering

18 worldwide (see Figure 4b). Weathering rates were generally higher in A0 throughout the

19 simulation, but it should be noted that in some areas in the final snapshot (year 12,000 CE)

weathering in A0 fell below that of B1. Since the only difference between B1 and B2 is the

presence of (globally averaged) atmospheric CO₂ concentration as a factor, the equivalent figure

for model version B2 (not shown) would have been extremely similar to B1.

3.3 Group C results

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The purpose of this group of experiments was to isolate and compare the individual contributions 1 2 of carbonate (C1) and silicate (C2) weathering to the global feedback mechanism, and to compare them with a scenario where this negative feedback does not exist (C3). The time series 3 of atmospheric CO₂ concentrations and DIC/alkalinity weathering are shown in Figure 8a. After 4 5 500 years of roughly similar behavior, the curves diverged into three distinct narratives. By year 6 12,000, about 33% of the emitted carbon was still in the atmosphere for model version C3 7 (constant weathering), whereas about 20% of the carbon remained for the C1 (change in carbonate weathering only), and 10% for C2 (change in silicate weathering only) and A0 (control 8 9 run). This is due to the immediate effect of carbonate weathering, which increase alkalinity 10 content in the ocean faster than the rate at which the precipitation of calcium carbonate increases. Over timescales of 10⁵ years or more, we would expect the C3 curve to catch up to C1 as 11 increased calcite burial releases carbon dioxide back to the ocean, negating the carbon removal at 12 the surface; this outcome is verified in the million-year simulations of Colbourn et al. (2013) but 13 impossible to replicate here due to the time scales involved. The C2 model version yielded very 14 similar results to A0, which included the impacts of both carbonate and silicate weathering. The 15 16 difference between the two was greater initially, as the additional alkalinity provided by carbonate weathering further enhances the oceanic uptake of CO₂ from the atmosphere, but the 17 18 gap gradually narrowed as the medium-term impacts of carbonate weathering faded away. 19 Again this outcome is verified over geologic timescales by Colbourn et al. (2013), with both the A0 and C2 equivalents returning the Earth system to pre-industrial levels. There was no change 20 in DIC weathering output from C2 since silicate weathering in this model does not increase the 21 22 DIC flux to the ocean. Alkalinity flux from C1 exceeded that of other model versions towards

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1 the end of the simulation period as the slower carbon removal resulted in much warmer surface

2 conditions compared to other model versions.

3 The evolution of ocean sedimentation is presented in Figure 8b using five model variables: ocean

4 carbon, calcite buried mass, upward/downward flux of calcite, and pore layer portion. Ocean

carbon levels remained similar between C1 and C2 during the first two thousand years, after

which C1 overtook C2 and eventually A0, despite resulting in the least amount of carbon

removal of the three model versions. This is because C1 would send as much DIC into the ocean

as alkalinity, which is counterproductive to atmospheric carbon removal. Inversely, C2 removed

9 almost as much carbon as A0 while adding 1000 Pg C less into the ocean, testifying towards the

efficiency of silicate weathering in removing carbon from the atmosphere compared to carbonate

11 weathering. Calcite sedimentation followed a very similar evolution to the experiments in Group

B (section 3.2), with the calcite pore layer portion remaining unchanged for 1000 years until the

upward flux of calcite (dissolution) became larger than the downward flux of calcite

(production/precipitation). Here it becomes clear that rock weathering, and in particular, silicate

15 weathering, is crucial in maintaining the stability of the pore layer fraction in the long term.

Model version C3, where weathering rates remain constant, produced a much sharper increase in

calcite dissolution compared to A0, where both weathering types respond to changes in climate,

and the burial of CaCO₃ in sediments occurred much faster in A0 than in C3. Additionally, pore

19 layer portion was better maintained by silicate weathering (C2) than carbonate weathering (C1).

These results suggest that the alkalinity flux supplied by silicate weathering is necessary not only

for decreasing the oceanic buffer factor (i.e., the concentration of carbonic acid and carbonate in

22 ocean surface water) and allowing the uptake of more CO₂ from the atmosphere, but also to

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1 maintain a better balance of the oceanic sedimentary pore layer by mitigating the increase in

2 calcite dissolution in the deep compared to the production rate in the surface layer.

3 4 Discussion

4 The weathering scheme introduced here is subject to some caveats relating to the formulation

itself, as well as the limitations inherent in the UVic model. Colbourn et al. (2013) carefully

discuss the potential misrepresentation of temperature as a factor, especially when other

parameters such as vegetation productivity are also taken into account. In particular, it is

8 possible that the temperature dependency for carbonate weathering (equation 7), which was

developed empirically from correlating groundwater CaCO₃ concentration with water

temperature in various river catchment basins, also captures the coincident changes in vegetation

productivity and river runoff, hence making the other factors redundant to a certain extent.

12 Whether this would introduce a significant error to the modeling is questionable, as temperature

on its own was shown to have at most a moderate impact on overall changes in weathering rates

14 (see Sect. 3.1).

15 The validity of the other two parameterizations – NPP and runoff – is difficult to assess as the

formulations are based on the arbitrary assumption that weathering rates vary monotonically and

linearly with changes in the two parameters. In the case of productivity dependence, for

18 example, the parameterization is meant to represent the physical impacts of root expansion, and

the chemical impacts of soil kinetics, on the breakdown of rock into minerals and their eventual

20 dissociation by carbonic acid. Thus an ideal productivity scheme would account for the impacts

of various plant types on each of the lithologies in terms of areal coverage, root expansion, and

22 other relevant quantities. Moreover, it should be noted that both the NPP and runoff schemes in

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1 our model rely heavily on the ratio between initial weathering and initial NPP/runoff, meaning

2 that a change in a parameter in an initially low-activity region (such as colder climates) may have

a disproportionately higher impact on weathering rates compared to changes in tropical areas. It

4 is possible that the introduction of vegetation in a previously nonvegetated area would introduce

a stress likely to drastically increase rock erosion, but this is an effect that would be better

represented by directly parameterizing the new plant type as a stress on the underlying lithology.

7 Therefore, a better parameterization may be one based on the absolute value of NPP/runoff

(using for example, a non-linear empirical function linking weathering rate and net primary

productivity) rather than the ratio of the current value to the initial value. The development of

such a relationship, however, would require a more in-depth investigation of the role of plants,

and biotic activity in general, on the physical and chemical erosion of rocks.

Another source of uncertainty in our results lies in the UVic model itself. While very well suited

to simulate long-term impacts of carbon emissions and increased weathering rates on ocean

biogeochemistry, on a shorter time scale (10²-10³ years) the lack of advanced atmospheric

dynamics prevents the model from adapting to the extreme warming brought on by carbon

emissions in a manner consistent with our understanding of global climate. Under extreme

warming there is a poleward shift of the tropical and subtropical cells and consequent changes in

18 precipitation patterns, leading to a potential overestimate of atmospheric temperature and

moisture content changes over tropical regions (see Sect. 3.1). This effect is important mostly

between years 2000-3000 CE, and fades away as the brunt of the climate and biogeochemical

changes are shifted to the oceans. The model's simplified precipitation scheme also likely

affects its ability to simulate runoff changes, which are central to both the initiation and

23 modulation of weathering rates.

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Terrestrial rock weathering is a complex mechanism with many variables worth considering, 1 2 many of which have a high degree of interdependence. In the scheme introduced in this paper for the UVic model, we considered the impacts of temperature, productivity and runoff (all 3 parameters previously examined in zero-dimensional weathering models), along with lithological 4 distribution to drive spatial variability. However, many other factors which affect weathering 5 rates were unaccounted for that could also be relevant in the context of a spatially explicit 6 7 weathering scheme. Perhaps the most meaningful of all is the consideration of sea level change. 8 It is highly likely that the extreme warming caused by anthropogenic emissions would result in a 9 significant melting of the Greenland and West Antarctic ice sheets (Clark et al., 2016), not only 10 disrupting the freshwater balance in the polar oceans, but also greatly contributing to a global rise in sea levels along with the thermal expansion of seawater. Many of the low-elevation 11 continental shelves threatened by sea level rise are situated in weathering active, tropical regions, 12 and therefore the interruption of terrestrial weathering due to the flooding of these areas could 13 substantially reduce the global weathering output, thus weakening the response to global 14 warming. Note that the extensive warming could also bring about a decrease in ice sheet area, 15 16 especially in Greenland, which would open up some potentially very active weathering regions (Kump and Alley, 1994). However, the extent of this areal reduction of ice sheet cover over a 17 18 few thousand years is likely to be overwhelmingly compensated by the area of land flooded by 19 sea level rise. 20 Another factor of some relevance is the interaction with land biogeochemistry. There has been 21 an extensive discussion in recent years on the role of mid- to high-latitude peatlands in the context of a rapidly warming Earth, especially with regards to the decarbonation of these 22 23 ecosystems and subsequent release of greenhouse gases in the atmosphere (mostly methane) that

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1 could greatly amplify global warming. While the release of methane by itself does not directly

2 affect terrestrial weathering, there are a variety of soil processes within peatlands which are

triggered or amplified by warming and which would have a significant local effect on the

4 chemical dissociation of rocks.

5 There are many other factors which would be worth investigating. For example, a distinction

6 between physical and chemical weathering would allow the inclusion of factors such as altitude,

as wind and relief/slope play a major role in physical weathering. The impact of ground frost at

8 higher latitudes also leads to erosion, and could increase weathering rates in colder climates.

9 Finally, one cannot ignore anthropogenic impacts, in particular the spread of modern agriculture,

in which crop yields are often boosted using mineral fertilisers and other chemicals, which mix

in with the soil water and accelerate the erosion of the bedrock. Other features of the

Anthropocene worth mentioning include acid rain and land use change, all of which need to be

taken into consideration in order to better represent the modern dynamics of global

biogeochemistry. Unfortunately, it is unlikely that most of these factors can be properly

integrated in current low- and intermediate-complexity climate models, on account of their

requiring a spatial resolution much finer than what most EMICs can offer. For example, the

UVic model's 1.8°×3.6° resolution cannot resolve physical mechanisms which occur over a

single-kilometer spatial scale.

5 Conclusions

20 A spatially-explicit weathering scheme has been developed and integrated into the University of

Victoria Earth System Climate Model (UVic ESCM). The model was constructed in such a way

that weathering rates at a certain point are computed based on the difference in temperature,

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1 vegetation net primary productivity, and runoff, between that point and pre-determined initial

2 conditions. In our model, those initial conditions were based on pre-industrial runoff and

lithology (Amiotte-Suchet et al., 2003), which provides the basis for the two-dimensionality of

4 the model.

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5 The model was tested with scenarios of future climate change, using (in most cases) a pulse of

5000 Pg C at year 2000 to simulate climate system recovery from anthropogenic emissions and

the role of global weathering during the following 10000 years. Overall, the model results

suggested that weathering has a negligible effect on atmospheric CO₂ and ocean biogeochemistry

on short timescales, but its impact becomes more discernible as we progress to multimillennial

timescales. We also found that climate system recovery from carbon emissions was much faster

using a two-dimensional model rather than the zero-dimensional model versions used in previous

work. Among the various climate factors used in the model, we found primary productivity to be

by far the most important, producing an increase in global weathering far higher than a model

version using atmospheric CO₂ levels to represent biotic activity, or one where only temperature

and runoff changes were considered. This highlights the need for further research to determine

whether this effect of biotic activity on physical and chemical weathering is in fact an important

real-world process that is independent of temperature and/or runoff change. Lithology itself was

also found to be very important, often meaning the difference between a weathering-active and

high- and low-weathering region. In terms of global totals, carbonate weathering was found to

be more prominent than silicate weathering, mostly on account of weathering-vulnerable rocks

being mostly carbonate-weathered. However, our results clearly emphasized that silicate

22 weathering is the only process of the two which has the capacity to fully restore the climate

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1 system to pre-industrial levels (on timescales of 10⁵ years), thus confirming the findings of

2 Colbourn et al. (2013).

3 This work has established the importance of using a spatially-explicit weathering scheme to

better represent long-term changes in carbon biogeochemistry. Our approach, although crude,

5 has demonstrated that weathering can be integrated on the grid-cell level and still produce

reasonable results. This study did not take into account the more subtle aspects of spatial

7 variability, such as the impacts of ice sheets, sea level changes, and local factors such as soil

8 activity and topography. These are therefore important processes to include in further analyses

of the effect of deglacial weathering changes on ocean biogeochemistry and climate change.

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Acknowledgements

12 We thank P. Amiotte-Suchet for providing the dataset for the worldwide rock type distribution

for the GEM-CO₂ model, as well as K. Meissner for providing the code for 0-D model versions,

14 A. Mucci for helping us better understand carbonate chemistry, and G. Colbourn for his

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Table captions

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Table 1. Rock type constants used in equations 5 and 6. Here, k represents the weathering rate

4 multiplier, with a higher multiplier signifying a higher concentration of weathering products per

5 unit of runoff (or alternatively, a lower resistance to weathering agents); α denotes the fraction of

the given rock type to weather as carbonate rocks. A value of zero means that the rock type

7 consists of silicate minerals only.

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9 **Table 2.** Description of each experiment carried out in this study. The emission total is the total

amount of carbon emitted by the pulse starting at year 2000, while the emission period represents

the time span of the pulse; the emission total is divided equally among the number of time steps

during the emission period. For the biological factor, "NPP" refers to equation 10, "Atm. CO₂"

to equation 11, and "None" signifies that this part of the weathering scheme is ignored. Finally,

when the CaCO₃ switch is OFF, the amount of carbonate weathering produced by the model is

set to its pre-industrial value for the duration of the simulation; similarly for when the CaSiO₃

switch is OFF.

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Figure captions

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3 Figure 1. Distribution of the six major rock types for the present day. Presented here are (a) the

4 source data from GEM-CO₂ [Amiotte Suchet et al., 2003] in 1°×1° resolution; (b) its adaptation

5 to the UVic model in 3.6°×1.8° resolution, displaying only the dominant lithology in each grid

6 cell; and (c) the interpolated rock type fraction in each grid cell. For the latter, the data is shown

7 ranging from 0 (white) to 1 (full color).

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9 Figure 2. Pre-industrial (year 1800 CE) setup for our weathering scheme. (a) Model simulated

annual mean river runoff, which is combined with rock type fractions (Figures 1 (b) and (c))

using equations 5 and 6 to produce (b) the carbonate and silicate weathering fluxes at pre-

industrial steady-state. Note the non-linear color scales, used here to better display values

13 outside of tropical regions.

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Figure 3. Time series of simulated changes in various globally-averaged or summed model

outputs for Group A scenarios, compared with pre-industrial steady-state values. These

scenarios include A0 (high-amplitude pulse), A1 (low-amplitude pulse), A2 (gradual emissions),

and their zero-dimensional counterparts (indicated by the "*" symbol). Shown here are (from

top to bottom): (a) atmospheric CO₂ concentrations, and weathering fluxes of DIC and alkalinity;

(b) global carbon budgets for atmospheric, ocean, land, and sediment reservoirs; and (c) surface

air temperature, net primary productivity, and oceanic concentrations of DIC and alkalinity.

Note the different scales along the time axis (separated by vertical dashed black lines). The

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1 curves shown here represent experiments A0 (red), A1 (blue), and A2 (green), with dashed

2 colored lines representing the zero-dimensional equivalent model version.

3

4 Figure 4. Spatial distribution of changes in (a) surface air temperature; (b) vegetation net

5 primary productivity; and (c) river runoff for experiment A0 from pre-industrial (year 1800 CE)

6 state to years 2100, 3000, 6000, and 12000 CE. Non-linear color scales are used in panels (b)

7 and (c) to better display the results for the later stages of the model simulation.

8

9 Figure 5. Spatial distribution of changes in carbonate (CaCO₃) and silicate (CaSiO₃)

weathering for experiment A0 from pre-industrial (year 1800 CE) steady-state to years 2100,

11 3000, 6000, and 12000 CE. Note the non-linear color scale, used to better display values during

the later stages of the model simulation.

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14 Figure 6. Time series of simulated changes in various model outputs for Group B scenarios,

15 compared with pre-industrial steady-state values. These scenarios include A0 (dependence on

temperature, NPP, and runoff), **B1** (dependence on temperature, atmospheric CO₂, and runoff),

B2 (dependence on temperature and runoff only), as well as their zero-dimensional counterparts

(indicated by the "*" symbol). Shown here are (from top to bottom): (a) atmospheric CO₂

concentrations, and weathering fluxes of DIC and alkalinity; and (b) oceanic carbon budget,

sediment carbon budget, downward flux of calcite into sediments, calcite pore layer portion, and

21 dissolution of calcite in sediments. Note the different scales along the time axis (separated by

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1 vertical dashed black lines). The curves shown here represent experiments A0 (red), B1 (blue),

2 and B2 (green), with dashed colored lines representing the zero-dimensional equivalent model

version.

4

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5 Figure 7. Spatial distribution of changes in carbonate (CaCO₃) and silicate (CaSiO₃) weathering

6 changes for experiment B1 between pre-industrial steady-state (year 1800 CE) and years 2100,

7 3000, 6000, and 12000 CE. Note the non-linear color scale, used to better display values during

8 the later stages of the model simulation.

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10 Figure 8. Time series of simulated changes in various model outputs for Group C scenarios,

11 compared with pre-industrial steady-state values. These scenarios include A0 (both weathering

types active), C1 (carbonate weathering only), C2 (silicate weathering only), C3 (no

weathering), as well as the zero-dimensional counterparts to C0 and C3 (indicated by the "*"

symbol). Shown here are (from top to bottom): (a) atmospheric CO₂ concentrations, and

weathering fluxes of DIC and alkalinity; and (b) oceanic carbon budget, sediment carbon budget,

downward flux of calcite into sediments, calcite pore layer portion, and dissolution of calcite in

sediments. Note the different scales along the time axis (separated by vertical dashed black

lines). The curves shown here represent experiments A0 (red), C1 (blue), C2 (green), and C3

(black), with dashed colored lines representing the zero-dimensional equivalent model version

20 (when available).

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1 Table 1

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Lithology	k	α	
Carbonate rocks	1.586	0.93	
Shales	0.627	0.39	
Sands and sandstones	0.152	0.48	
Basalts	0.479	0	
Shield rocks	0.095	0	
Acid volcanic rocks	0.222	0	

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1 Table 2

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Group	Experiment Name	Emission total (Pg C)	Emission period (years CE)	Biological parameter	CaCO ₃ weathering switch	CaSiO ₃ weathering switch
A	A0	5000	2000-2001	NPP	ON	ON
	A1	1000	2000-2001	NPP	ON	ON
	A2	5000	2000-3000	NPP	ON	ON
В	B1	5000	2000-2001	Atm. CO ₂	ON	ON
	B2	5000	2000-2001	None	ON	ON
С	C1	5000	2000-2001	NPP	ON	OFF
	C2	5000	2000-2001	NPP	OFF	ON
	СЗ	5000	2000-2001	NPP	OFF	OFF

Note: The "*" notation refers to zero-dimensional versions of the model using otherwise

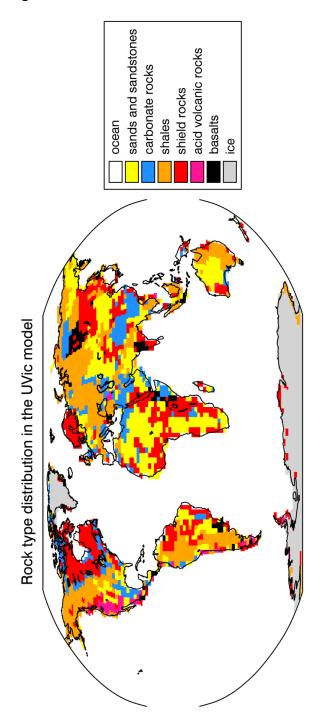
⁴ identical experimental parameters.

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1 Figure 1a

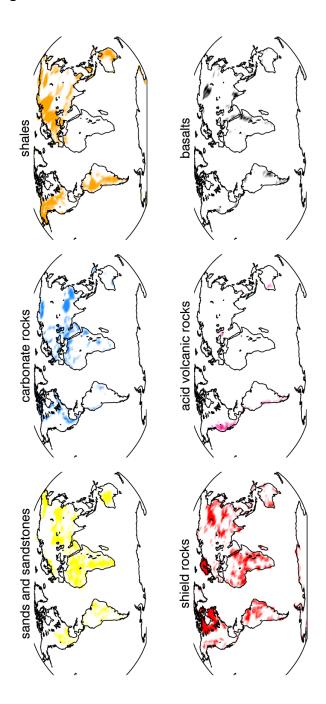


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Figure 1b

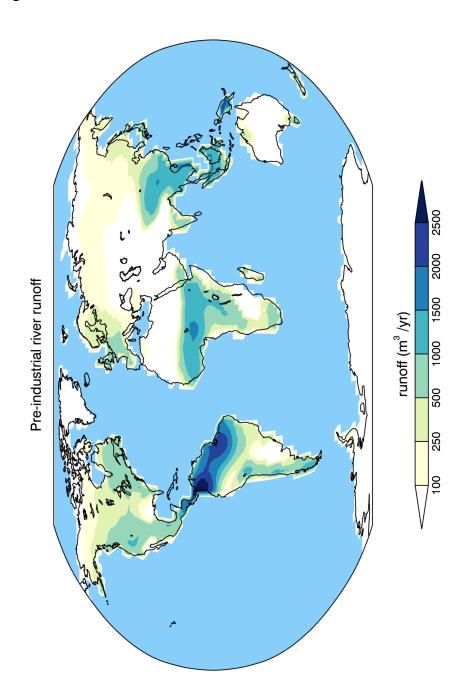


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1 Figure 2a

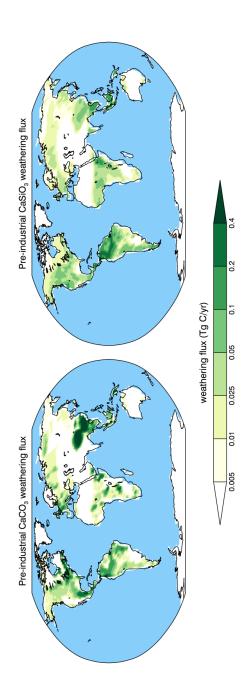


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Figure 2b



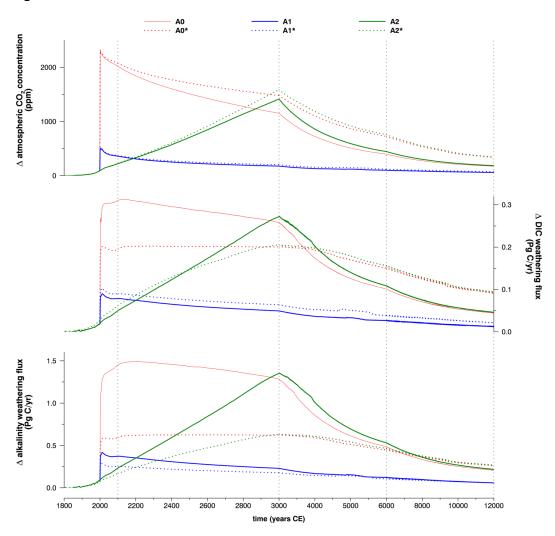
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Figure 3a

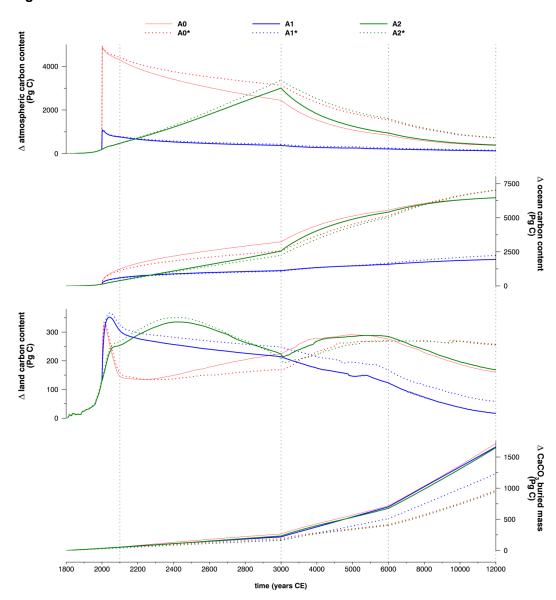


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Figure 3b

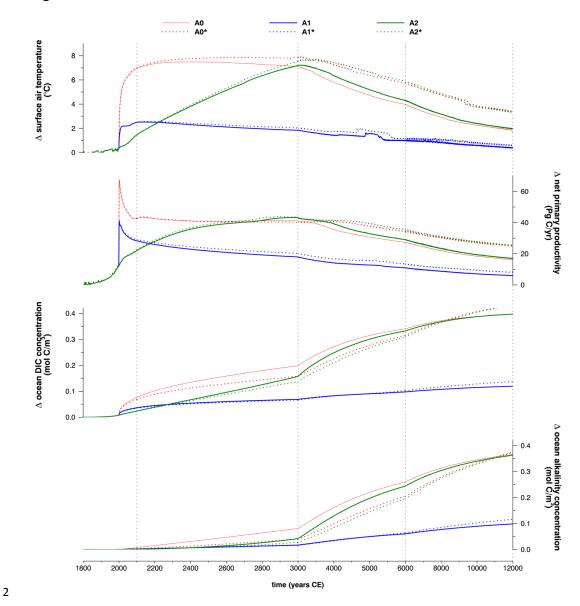


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1 Figure 3c

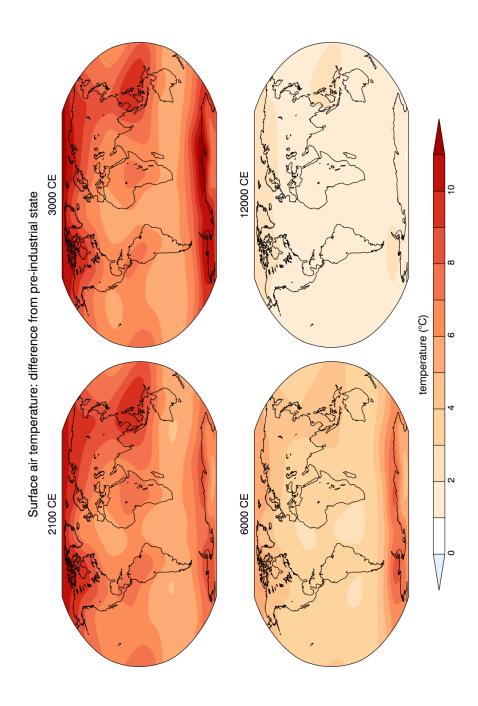


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Figure 4a

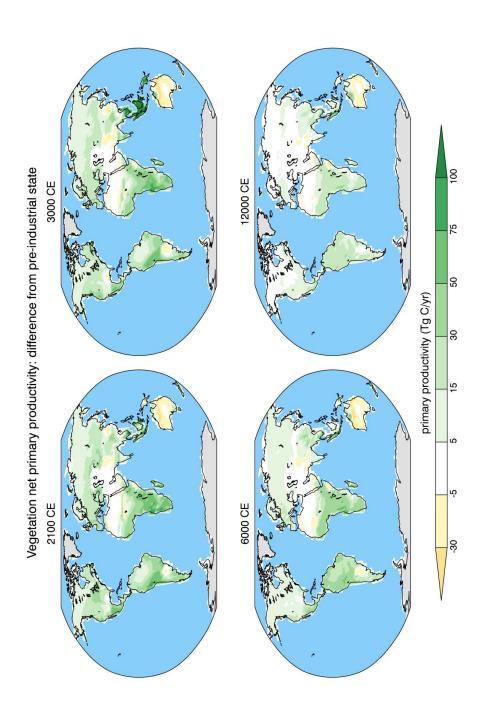


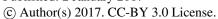
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1 Figure 4b

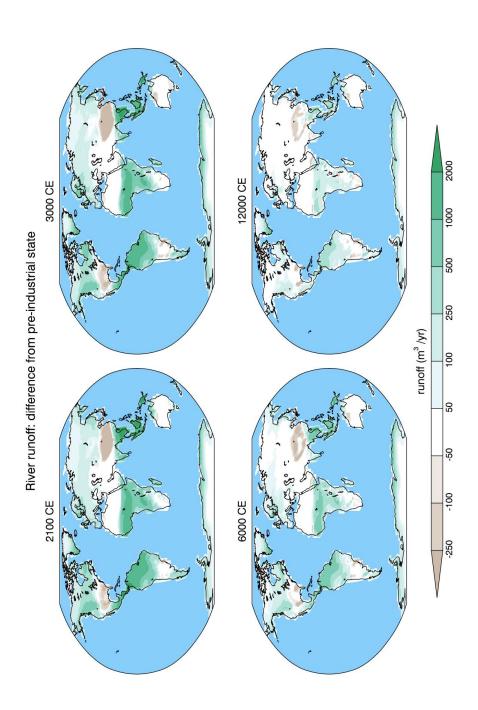








1 Figure 4c



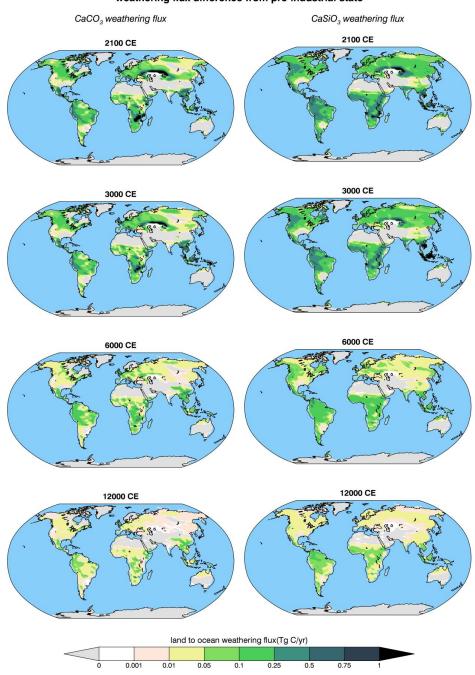
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1 Figure 5

weathering flux difference from pre-industrial state

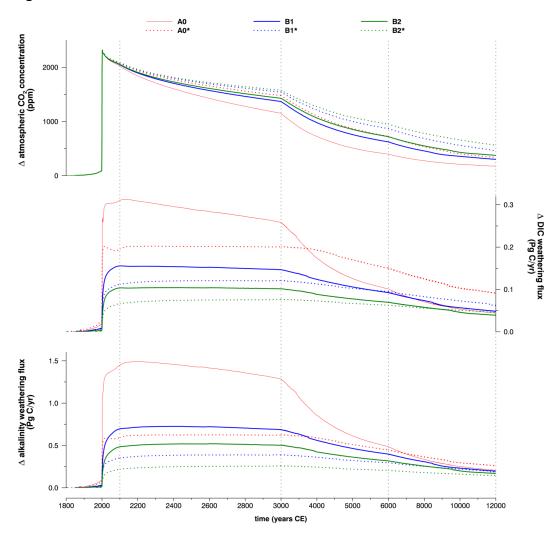


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1 Figure 6a

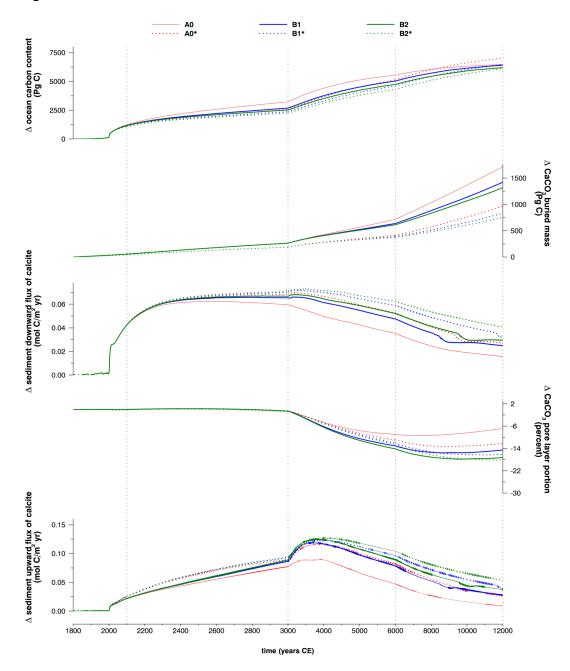


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1 Figure 6b



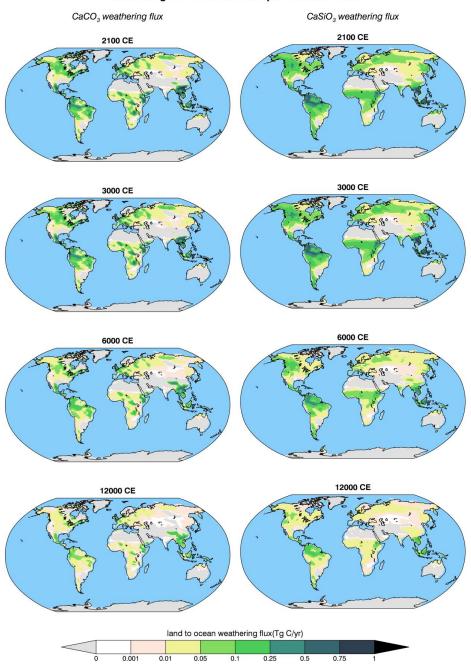
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1 Figure 7

weathering flux difference from pre-industrial state

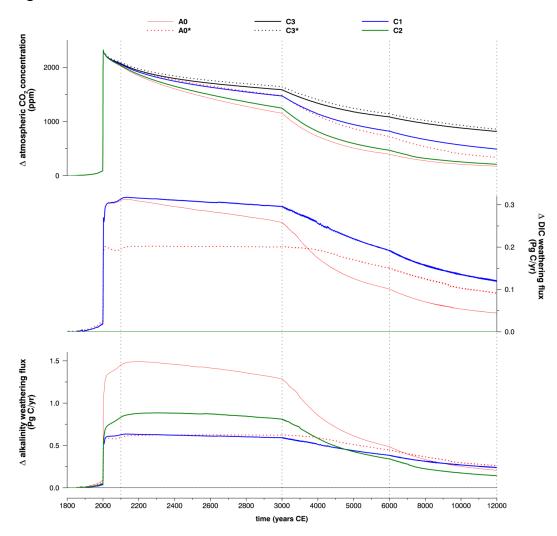


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1 Figure 8a



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1 Figure 8b

