

1 Mountains, erosion and the carbon cycle

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7

8 **Abstract**

9 Mountain building results in high erosion rates and the interaction of rocks with the
10 atmosphere, water and life. Carbon transfers that result from increased erosion could control
11 the evolution of Earth's long-term climate. For decades, attention has focused on the
12 hypothesised role of mountain building in drawing down atmospheric carbon dioxide (CO₂)
13 via silicate weathering. However, it is now recognized that mountain building and erosion
14 affect the carbon cycle in other important ways. For example, erosion mobilises organic
15 carbon (OC) from terrestrial vegetation, transferring it to rivers and sediments and thereby
16 acting to draw down atmospheric CO₂ in tandem with silicate weathering. Meanwhile,
17 exhumation of sedimentary rocks can release CO₂ through the oxidation of rock OC and
18 sulfide minerals. In this Review we examine the mechanisms of carbon exchange between
19 rocks and the atmosphere and discuss the balance of CO₂ sources and sinks. Our Review
20 demonstrates that OC burial and oxidative weathering, not widely considered in most models,
21 control the net CO₂ budget associated with erosion. Therefore, lithology strongly influences
22 the impact of mountain building on the global carbon cycle, with an orogeny dominated by
23 sedimentary rocks, and thus abundant rock OC and sulfides, tending towards being a CO₂
24 source.

25 **1. Introduction**

26 Mountain building and the production of topography, owing to the upward migration
27 of rock following plate convergence or dynamic uplift, is accompanied by increases in
28 physical erosion rates¹, rock exhumation²⁻⁵, and sediment transfer by rivers to the oceans^{6,7}.
29 In 1899, Chamberlin hypothesised that heightened erosion associated with mountain building

30 would increase the draw down of carbon dioxide (CO₂) from the atmosphere by facilitating
31 chemical weathering of silicate minerals⁸. Ninety years later Chamberlin's idea blossomed,
32 reinvigorated by records of changes in ocean chemistry⁹ that linked uplift of the Himalaya
33 and Tibet¹⁰ with global cooling over the Cenozoic^{11,12,13}. A vigorous debate regarding the role
34 of Himalayan uplift in driving weathering and CO₂ drawdown ensued¹⁴⁻¹⁷, stimulating
35 research that sought to better constrain the controls on silicate weathering rates in the
36 field^{18,19} and laboratory²⁰.

37 The premise of a coupling between erosion, weathering, and the carbon (C) cycle is
38 based on the reaction of silicate minerals with carbonic acid (Box 1, Eq. 1). Chemical
39 weathering of silicate minerals has long been considered to be a key mechanism for removing
40 atmospheric CO₂, counterbalancing the release of carbon from volcanism and, therefore,
41 maintaining a habitable planet²¹⁻²⁴ (Fig. 1A). Nevertheless, despite work extending back to
42 the 19th Century, quantitative understanding of how chemical weathering responds to uplift
43 and erosion, and the associated consequences for the global C cycle, remained limited even in
44 the 1990s.

45 Over the past three decades, several advances have enabled an improved
46 understanding of the effect of erosion on silicate weathering. For example, geomorphological
47 frameworks describe the erosional and physical weathering processes that expose fresh
48 mineral surface area²⁵. In addition, geochemical kinetics²⁶ and reactive transport
49 modelling^{27,28}, informed by intensive field data collected from critical zone observatories²⁹⁻³¹,
50 quantify how rates of weathering reactions vary in response to mineral surface exposure as
51 well as to hydrological controls²⁰. As a result, the global effect of uplift and erosion in
52 enhancing interaction between minerals and the atmosphere and hydrosphere can now be
53 quantified³²⁻³⁴.

54 Over the same time period, it has become clear that understanding the net effect of
55 erosion on the C cycle requires looking beyond silicate weathering³⁵ (Fig. 1B). In this
56 emerging view, erosion both drives the transport and burial of organic carbon (OC) in ocean
57 sediments (an additional CO₂ sink)³⁶⁻³⁹ and exposes rock-derived OC and minerals that can
58 be oxidised during weathering (CO₂ source; Box 1). Specifically, both oxidation of rock-
59 derived, or "petrogenic," OC (OC_{petro})^{40,41} and oxidation of sulfide minerals to produce
60 sulfuric acid, which can, in turn, react with carbonate minerals^{42,43}, act as CO₂ sources (Box
61 1, Fig. 1B). To fully understand the role of erosion in the C cycle, all of the mechanisms that

62 transfer C between rocks and the atmosphere must be considered, including both CO₂ sinks
63 (silicate weathering and OC burial) and CO₂ sources (OC_{petro} and sulfide oxidation).

64 In this Review, we examine the various processes that lead to transfer of C between
65 rocks and the atmosphere, focusing on how these relate to erosion. We examine the
66 functional relationships between erosion rate and carbon fluxes, focusing on river catchment-
67 scale measurements from the modern day. Our Review considers components of the C cycle
68 which have remained poorly constrained until recently and, using a holistic approach that
69 considers the inorganic and organic carbon cycles, we provide a new framework to identify
70 whether mountains are a net source or sink of CO₂.

71 **2. The geological carbon cycle**

72 Solid Earth CO₂ degassing, associated with volcanic arcs, intraplate volcanoes and
73 mid-ocean ridges, contributes ~70-100 megatonnes C per year (MtC yr⁻¹) to the Earth's
74 atmosphere⁴⁴. Additional CO₂ contributions from diffuse continental rifts⁴⁵ and
75 metamorphism^{46,47} are poorly constrained, but could be similar in magnitude.

76 The pre-industrial C content^{48,49} in Earth's surface reservoirs (atmosphere, oceans and
77 biosphere) was $43,542 \pm 550 \times 10^3$ MtC. Although the annual degassing flux may seem small
78 compared to this mass, it would take <500,000 years for degassing (at ~100 MtC yr⁻¹) to
79 replace the entire inventory of C in the surface reservoirs — a relatively short time in the
80 geological context. On timescales from years to thousands of years, the evolution of
81 atmospheric CO₂ is governed by the substantially larger, and closely balanced, C fluxes
82 associated with photosynthesis and respiration, and ocean-atmosphere gas exchange (Fig. 2).
83 As a result, without counteracting geological sinks, CO₂ released from solid Earth degassing
84 would accumulate over million-year timespans to levels that are unrealistically high¹⁷. In the
85 “textbook” view of the C cycle, the draw down of CO₂ by silicate weathering acts as the
86 counteracting sink, and the response of weathering to erosion provides a mechanism for
87 driving changes in atmospheric CO₂.

88 **2.1 The textbook view of the C cycle**

89 Chemical weathering of silicate minerals has long been recognized as a key process
90 causing the long-term draw down of atmospheric CO₂ (ref. ^{21,50}; Box 1). Modern-day CO₂
91 drawdown by silicate weathering is ~90-140 MtC yr⁻¹, similar in magnitude to the flux of

92 CO₂ released from solid Earth degassing (Fig. 1A)^{18,51}, with the balance between these fluxes
93 controlling atmospheric CO₂ and climate in textbook C cycle models. Climatic factors such
94 as temperature, precipitation, and biological productivity (which produces high
95 concentrations of CO₂ in soils and organic acids that drive weathering), influence the flux of
96 CO₂ drawdown from silicate weathering (Box 1). The climate-dependency of weathering
97 produces a negative feedback between atmospheric CO₂ and global climate^{23,24} such that
98 imbalances in atmospheric CO₂, for example, driven by changes in solid Earth CO₂
99 degassing, are balanced by changes in weathering over millions of years¹⁷.

100 Within this textbook framework, changes in the reactivity of silicate minerals, for
101 example, caused by mountain uplift and erosion, can drive global cooling by lowering
102 atmospheric CO₂ concentrations. Changes in the atmospheric CO₂ concentrations might,
103 subsequently, alter the weathering flux, maintaining the overall balance of the carbon cycle
104 and establishing a new steady-state climate regime^{52,53}. Such effects are known as the
105 “weathering thermostat” and are regularly included in canonical carbon cycle models such as
106 GEOCARB⁵⁴ and COPSE⁵⁵.

107 Critical to the weathering thermostat paradigm is the response of weathering fluxes to
108 changes in CO₂ concentrations in the atmosphere. Low-lying continental terrains, however,
109 might not be able to provide a sufficient chemical weathering flux to sustain a feedback
110 response^{32,56}, leading to non-steady-state behaviour in the C cycle. Such limits to the climate
111 stabilization were rarely, if ever, exceeded in the Phanerozoic⁵⁷ or even earlier in Earth’s
112 history. Although the mechanism by which erosion might drive climate remains debated –
113 and will be our focus throughout this Review – canonical C cycle frameworks inherently
114 require mountain building and erosion to have played a key role by sustaining the global
115 silicate weathering feedback.

116 ***2.2 An emerging view of the C cycle***

117 The textbook juxtaposition of C sources from solid Earth degassing versus C sinks
118 from silicate weathering has prevailed for decades^{17,24} and persists today^{53,58} (Fig. 1A). Yet,
119 other C fluxes are clearly important to the geological C cycle^{36,39,41}. In this Review, we
120 advocate for a more holistic view of the geological C cycle that considers CO₂ drawdown via
121 organic carbon burial^{37,39,59}, as well as CO₂ sources from OC_{petro} oxidation^{40,41} and sulfide
122 oxidation coupled to carbonate weathering^{42,43}. The global fluxes associated with these

123 processes (Fig. 1B) are similar in magnitude to those from volcanism and silicate weathering
124 and, critically, have strong links to erosion. Therefore, we argue that the role of mountain
125 building in the C cycle cannot be understood by considering silicate weathering in isolation.
126 Instead, we must address how uplift and erosion affect each of the key transfers of C between
127 rocks and the atmosphere.

128 In the following section, we assess each process of C transfer between rocks and the
129 atmosphere, including their relative fluxes and how they are linked to erosion and mountain
130 building. We then evaluate the net “rock-atmosphere” budget for multiple locations, which
131 allows us to understand the cumulative effect of erosion and mountain building on the carbon
132 cycle. If the net rock-atmosphere transfer increases in response to erosion (for example, if
133 erosion increases the relative importance of weathering-related sources of CO₂ over the CO₂
134 sinks), there should be a transient surplus of CO₂ associated with mountain building,
135 warming the planet and restoring C cycle equilibrium via negative feedbacks. If the net rock-
136 atmosphere transfer decreases in response to erosion, the overall effect of mountain building
137 could be removal of CO₂ from the atmosphere and global cooling^{8,12}.

138 The wider perspective we propose is also relevant to the cycle of dioxygen (O₂).
139 While not the focus of this Review, the OC cycle and oxidative weathering of sulfides also
140 affect the budget of atmospheric O₂ (refs.^{60,61}). Atmospheric O₂ concentrations have been
141 relatively high throughout the Phanerozoic, so perturbations to OC and sulfur cycles, which
142 could impact atmospheric CO₂, are unlikely to have had a substantial impact on atmospheric
143 O₂ concentrations (refs.^{55,60,62}). That said, some perturbations to atmospheric O₂ might be
144 detectable in Pleistocene ice cores⁶³. Earlier in Earth’s history, links between atmospheric
145 CO₂, O₂ and climate are intriguing^{61,64} and the role of erosion and weathering is worthy of
146 further attention.

147 **3. Mountains, erosion and carbon transfer**

148 As mountain building takes place over millions of years, and erosion rates respond
149 over timescales of thousands of years or more, it is not possible to observe their effects on C
150 cycling directly. Instead, we can estimate CO₂ sources and sinks in modern environments
151 and, thus, examine how the CO₂ flux associated with each C transfer mechanism varies
152 across a range of erosion rates. Specifically, the chemistry of soils and rivers can be used to

153 examine how spatial differences in erosion rate, climate (temperature and precipitation) and
154 lithology link to C transfers and fluxes in different regions.

155 The amount and composition of material transported in rivers provides important
156 information about the fluxes of carbon delivered to the oceans, where most long-term C
157 burial in carbonate and organic matter occurs. Although river fluxes are a remarkable
158 resource for quantifying C fluxes, they are biogeochemically active and thus more than
159 simple conduits for C. For example, substantial amounts of C younger than a few decades old
160 are lost as CO₂ by active degassing from rivers^{65,66}. The degassing fluxes associated with
161 biogeochemical activity in rivers, while not the focus of this Review and generally not
162 important for assessing long-term C sources and sinks, are relevant for the short-term C cycle
163 (Fig. 2).

164 ***3.1 Erosion and carbon sinks***

165 In this subsection, we discuss the various processes by which erosion might act to draw down
166 CO₂ from the atmosphere, and how the fluxes associated with these processes vary as a
167 function of erosion rate in modern environments.

168 3.1.1 Erosion and silicate weathering.

169 Silicate weathering can be tracked by measuring the flux of the dissolved cations
170 (such as Ca²⁺, Mg²⁺, Na⁺, K⁺) transported as solutes in rivers, and accounting for cation
171 sources other than silicate minerals (such as carbonate and evaporite minerals, atmospheric
172 deposition, and anthropogenic inputs)^{18,19,67}. Data from modern rivers suggest that ~40% of
173 global chemical denudation occurs on the steepest 10% of the Earth's surface⁶⁸. As a result,
174 mountainous regions appear to have a large influence on the total fluxes from chemical
175 weathering. However, understanding the effects of weathering on the C cycle depends on
176 distinguishing silicate weathering (which drives CO₂ drawdown) from carbonate weathering
177 (which does not). The proportion of solutes derived from carbonate versus silicate weathering
178 generally increases with erosion rate⁶⁹. Therefore, though there is an apparent linear
179 relationship between erosion rate and total weathering fluxes⁶⁸, this relationship might not
180 reflect an erosional control on long-term CO₂ drawdown. Geochemical mixing models can be
181 used to correct for carbonate sources^{18,32,69} and evaluate the true relationship between silicate
182 weathering and erosion. Once focusing solely on silicate weathering, substantial scatter
183 emerges in the relationship with denudation rate (Fig. 3a).

184 The scatter between physical denudation rates and silicate weathering flux can be
185 understood by considering how weathering fluxes depend on the interplay between the supply
186 of material by erosion and the speed of chemical reactions. At low erosion rates, weathering
187 products accumulate in soils and shield underlying rocks from interactions with surficial
188 fluids, leading to “supply limited” weathering^{18,32,33,70-72} (the term “transport limited” has
189 also been used to describe this phenomenon; Fig. 3a). Under supply limited conditions,
190 weathering and erosion scale linearly (Fig. 3a), as increased erosion rates expose more
191 material for reaction^{32,33,73}. In contrast, at higher erosion rates (Fig. 3a), minerals are supplied
192 in excess of the rate at which they react. Fluxes are then “weathering limited” (also referred
193 to as “kinetically limited”) and are controlled by other factors, such as CO₂ concentration,
194 fluid flow rates, and temperature, so that they depend only weakly (if at all) on erosion rate<sup>32-
195 34,73,74</sup>.

196 In supply limited weathering, silicate weathering fluxes are less sensitive to climate
197 compared to weathering limited regions and provide a weak (or potentially inactive) feedback
198 to temperature and hydrological change. By contrast, the climate-silicate weathering feedback
199 is strongest in areas that are characterised by high erosion rates^{32,34,74}. Therefore, mountain
200 building and erosion play key roles in the evolution of atmospheric CO₂ by modulating CO₂
201 drawdown as well as the sensitivity of CO₂ removal fluxes to climatic changes (ref.⁷⁵).

202 3.1.2. Erosion and burial of organic carbon.

203 Geomorphic processes such as landslides and overland flows erode terrestrial organic
204 matter from plants and soils. The terrestrial organic matter (commonly referred to as
205 biospheric OC; OC_{biosphere}) is then exported as part of the solid load of rivers^{76,77}. If the
206 OC_{biosphere} is buried in sediments and escapes subsequent degradation, and new plant growth
207 replaces the organic matter at the site of erosion, the overall effect is to draw down
208 atmospheric CO₂ (refs.^{78,79}). Globally, erosion of particulate OC_{biosphere} represents a sizable
209 and dynamic geological CO₂ sink (Fig. 1B, Box 1, Eq. 5)^{36,50,80,81}. The total OC burial flux in
210 modern ocean sediments^{82,83} is ~170 MtC yr⁻¹, of which ~40-80 MtC yr⁻¹ is estimated to be
211 derived from OC eroded from land^{39,84}, facilitated by higher preservation efficiency (10-30%)
212 compared to marine organic matter (<1.3%)^{38,85}.

213 We can explore the empirical relationship between erosion rate and OC_{biosphere} fluxes
214 by measuring the flux of C carried by rivers. However, first we need to isolate the relative

215 contribution of OC_{biosphere} and OC_{petro} to the measured C flux in the river solid load, which is
216 typically achieved using analysis of radiocarbon (¹⁴C)^{86,87}. Radiocarbon can distinguish
217 sources of OC in river sediments as OC_{petro} is radiocarbon-dead (low ¹⁴C activity), while
218 OC_{biosphere} is not (high ¹⁴C activity)^{88,89}. Therefore, using this approach, the global flux of
219 OC_{biosphere} carried by rivers to the oceans can be calculated, yielding a value of 157^{+74/-50}
220 MtC yr⁻¹ (ref.⁹⁰). Steep topography (> 10° slope angles) is important in driving this flux^{59,91},
221 contributing ~20-40% to the global total⁹². In addition, considering C fluxes across
222 catchments that display a range in erosion rates reveals a broad positive relationship between
223 OC_{biosphere} export and suspended sediment yield (Fig. 3b)^{90,93}.

224 The observed sub-linear scaling between the suspended sediment yield and OC_{biosphere}
225 export indicates that there is a lower fractional contribution of OC_{biosphere} to river sediments in
226 areas with higher erosion rates, can be explained by a shift from shallow erosion (mobilizing
227 OC-rich material in soils) to deeper erosion characterized by landslide activity^{94,95} (Fig. 3b).
228 As a result, OC_{biosphere} export is less efficient with increasing erosion⁹⁰ — potentially putting
229 a limit on erosional forcing of this carbon sink. However, higher erosion rates might increase
230 the likelihood that terrestrial OC_{biosphere} is buried and preserved in sediments^{39,59}. Associations
231 between OC_{biosphere} and mineral surfaces can protect organic compounds from degradation^{82,96}
232 regardless of their inherent reactivity⁹⁷. In addition, high sedimentation rates can bury organic
233 matter more rapidly, thereby decreasing contact with oxygenated seawater and increasing
234 burial efficiency of OC_{biosphere} (ref.⁹⁸). Preservation of OC_{biosphere} associated with increased
235 burial and/or associations with mineral surfaces enhance the links between erosion and draw
236 down of CO₂ by OC_{biosphere} burial.

237 Climate can moderate the flux of OC_{biosphere} that results from increased erosion. For
238 example, large storms can trigger landslides and drive river sediment transport. As such,
239 OC_{biosphere} transfer during intense rainfall events is a common feature of mountain rivers in
240 Taiwan^{86,93,99}, the Andes^{87,100}, North America^{101,102}, the Longmen Shan of eastern Tibet¹⁰³,
241 and the European Alps¹⁰⁴. Storms can also mobilise OC in the form of coarse woody
242 debris^{105,106}, and temporal correspondence of high sediment and OC_{biosphere} fluxes during
243 floods⁸⁶ could increase the chance of long-term burial⁵⁹. As a result, a warm and wet climate
244 can enhance OC_{biosphere} export and burial. By doing so, erosion of OC_{biosphere} could act as a
245 negative feedback on geological CO₂ emissions, working in tandem with silicate
246 weathering⁹².

247 3.1.3. Erosion and supply of rock-borne nutrients

248 Erosion and weathering can control the supply of critical rock-derived nutrients to both the
249 terrestrial and marine biosphere. On one hand, in terrestrial ecosystems, erosion and
250 weathering can influence the C stocks of forests by modulating the supply of phosphorus (P),
251 calcium (Ca) and potentially nitrogen¹⁰⁷⁻¹⁰⁹. On the other hand, erosion can also facilitate
252 nutrient loss^{110,111}, and steep slopes can reduce soil thickness and impact water availability,
253 limiting productivity¹¹². As a result, the direct impact of erosion on forest carbon stocks is
254 complex. We do know, however, that erosional export of OC_{biosphere} is not limited by net
255 primary productivity^{90,93} — so while erosion-enhanced terrestrial productivity might affect
256 forest carbon stocks on land, it is not expected to influence long-term transfer of C into the
257 rock reservoir (Fig. 2). Meanwhile, weathering also influences the supply of limiting
258 nutrients to the oceans. For example, delivery of P can drive marine productivity and OC
259 delivery to marine sediments^{38,82}. Some biogeochemical models incorporate weathering-
260 dependent delivery of P to the oceans^{64,113}, but little is known about the ways in which
261 erosion controls this flux — reflecting a key knowledge gap and an opportunity for further
262 work.

263 ***3.2 Erosion and carbon sources***

264 In this subsection, we discuss how, in addition to the CO₂ sinks described above, erosion can
265 lead to C release from rocks to the atmosphere (as CO₂) and consider how the resulting fluxes
266 are linked, in important ways, to erosion rates.

267 3.2.1 Rock organic carbon oxidation.

268 The upper 1 m of Earth's surface contains an estimated $\sim 10^6$ MtC of OC_{petro}, which is
269 often millions of years old^{114,115}. When this OC_{petro} is brought towards the surface during
270 exhumation of the bedrock, some of it is oxidised, releasing CO₂ to the atmosphere.
271 Oxidation can occur in the upper centimetres to meters of the Earth's surface, where active
272 weathering takes place via interaction with air and O₂-bearing water¹¹⁶⁻¹¹⁸, or in sediments
273 transiting large river floodplains^{119,120}. Altogether, a global “geo-respiration” flux of ~ 40 -100
274 MtC yr⁻¹ occurs through OC_{petro} oxidation⁴⁰ (Box 1, Eq. 6).

275 The flux of oxidised OC_{petro} might be closely linked to erosion⁴¹. In the presence of
276 O₂-rich atmosphere and surface waters, OC_{petro} oxidation occurs at a faster rate than chemical

277 weathering of silicate minerals^{121,122}, in principle allowing oxidative weathering to keep pace
278 with increased supply of material by erosion¹²³. However, quantifying the rate of OC_{petro}
279 oxidation across different landscapes is challenging, so historically it has been difficult to test
280 this prediction. Recently, riverine dissolved fluxes of rhenium, an element that is associated
281 with OC_{petro} in rocks, have been used as a proxy to measure the weathering (and presumably
282 subsequent oxidation) of OC_{petro}^{41,124}. Resulting data from sedimentary lithologies, which
283 contain OC_{petro}, confirm that erosion rate exerts a first order control on OC_{petro} oxidation (Fig.
284 3c).

285 The observed scatter between OC_{petro} oxidation rate and suspended sediment yield
286 could relate, in part, to the variable OC_{petro} content of rocks undergoing weathering (Fig. 3c).
287 However, not all exhumed OC_{petro} is weathered^{89,125,126}, and the proportion that is lost by
288 oxidation ranges from ~10%-40% across mountain catchments^{41,127} up to ~70% in mountain
289 soils¹²⁸ and >90% in large tropical rivers¹²⁰. Some of the unoxidised OC_{petro} observed in
290 many river sediments could reflect a chemically and physically resilient component¹¹⁹ that
291 formed through interactions between organic matter and minerals^{96,97} and/or during high
292 temperature metamorphism¹²⁹. For instance, graphitic OC_{petro} can persist through large
293 tropical floodplains^{119,120}, and over several cycles of mountain building, erosion, deposition
294 and exhumation¹³⁰. Alternatively, the presence of un-weathered OC_{petro} in river sediments
295 might indicate that OC_{petro} oxidation is locally limited by temperature, O₂ supply, and/or
296 microbial activity^{121,123,128,131}, especially at high erosion rates. Therefore, climatic factors
297 might be linked to the rate of CO₂ emission via oxidation of OC_{petro} (Fig. 1B), a hypothesis
298 that could be tested using direct measurements of CO₂ release during oxidative weathering¹¹⁸.
299 Direct measurements also have the advantage of measuring CO₂ flux itself, rather than
300 tracking the loss of OC_{petro} and assuming that all of the lost carbon is eventually respired⁴¹.

301 3.2.2. Sulfide oxidation.

302 In addition to OC_{petro} oxidation, sulfide oxidation provides a potential CO₂ source.
303 When sulfide minerals such as pyrite are exhumed, they are oxidized and produce sulfuric
304 acid (H₂SO₄). If this acid reacts with carbonate minerals or alters the carbonic acid
305 equilibrium in water, the net effect can be CO₂ release to the atmosphere^{19,42,43,118,132} (Box 1,
306 Eq. 3&4). The amount of sulfide weathering can be calculated from riverine sulfate (SO₄²⁻)
307 concentrations and fluxes. The effect of sulfide weathering on CO₂ fluxes can then be
308 inferred in the context of other weathering reactions determined by dissolved ion

309 chemistry¹³². However, in addition to being the product of sulfide oxidation, dissolved
310 riverine sulfate can also originate from anthropogenic and/or evaporite mineral sources¹⁸. The
311 stable isotope composition of S and O in dissolved sulfate can distinguish the relative
312 contribution of each sulfate source^{42,132-135}(Fig. 3d).

313 Measurement of sulfide fluxes and S and O isotope analyses suggest substantial
314 global CO₂ release associated with sulphide oxidation (~40 MtC yr⁻¹), although this estimate
315 remains poorly constrained^{132,133}. Data from river catchments show a broad positive
316 correlation between pyrite oxidation and erosion rate⁴², confirming that erosion has a first
317 order control on the sulfide-related flux of CO₂ (Fig. 3d) and suggesting that pyrite oxidation
318 is generally a supply-limited process. The relationship between pyrite oxidation and erosion
319 rate is not surprising since, similar to OC_{petro}, pyrite oxidation occurs rapidly^{122,136} and so is
320 expected to increase with erosional supply. As a consequence, mountain building could
321 enhance the release of CO₂ by this mechanism.

322 The effects of sulfide oxidation are reversed when SO₄²⁻ is removed from seawater,
323 typically by reduction and formation of sulfide minerals in marine sediments¹³⁷. However,
324 SO₄²⁻ has a long residence time in the oceans (~10 Myrs), so increases in global fluxes from
325 sulfide oxidation can sustain elevated atmospheric CO₂ concentrations over million-year
326 timescales before the SO₄²⁻ is removed⁴³. Thus, changes in C fluxes associated with sulfide
327 oxidation, for example driven by erosion, can be important for the long-term C cycle.
328 Additional processes in river systems and marine sediments, including coupling of sulfide
329 oxidation with carbonate vs. silicate mineral weathering¹³⁸, as well as the effect of Fe-S redox
330 cycling on sulfur budgets¹³⁷, might play important roles in modulating the net effect of
331 sulfide oxidation on C transfer fluxes and thus deserve more attention in future work.

332 **3.3 Net ‘rock-atmosphere’ carbon transfers**

333 We have reviewed emerging datasets that reveal an erosional control not just on
334 silicate weathering, but also on other fluxes in the geologic C cycle such as sulfide and
335 OC_{petro} oxidation (Fig. 3). Considered together, present day estimates of the fluxes associated
336 with each “rock-atmosphere” C transfer mechanism indicate that the global sink of
337 atmospheric CO₂, via silicate weathering and OC burial, is ~175-240 MtC yr⁻¹ (Fig. 1B).
338 Conversely, the release of CO₂ from sedimentary rock weathering and volcanic degassing is
339 40-140 MtC yr⁻¹ and 70-100 MtC yr⁻¹, respectively. Combined, these CO₂ sources total 110-

340 240 MtC yr⁻¹, similar in magnitude to the CO₂ sink from silicate weathering and OC burial.
341 This CO₂ budget, however, does not account for poorly constrained fluxes from CO₂
342 outgassing in continental rifts⁴⁵ or metamorphic processes^{46,47}.

343 The large uncertainties surrounding the CO₂ source and sink fluxes indicate that
344 further work is required to define these values more precisely. Nevertheless, the similar
345 magnitude of the geological sources and sinks could keep the C cycle in an approximate
346 steady-state balance over million-year timescales (Fig. 2). Within this overall budget there is
347 scope for small, transient imbalances that could drive shifts in global climate. The
348 geochemical thermostat (increasing CO₂ drawdown and C burial in response to rising
349 atmospheric CO₂, or vice versa) associated with silicate weathering and/or OC_{biosphere} burial,
350 would then act to return C fluxes to a new steady state (and corresponding changes in global
351 climate). Mountain building could potentially drive such transient C cycle perturbations and
352 associated climatic change.

353 Insights into how C cycle imbalances can arise comes from river catchments where
354 enough data have been collected to quantify each rock-atmosphere C transfer term (Fig. 1B,
355 and Supplementary Information): four catchments in the western Southern Alps of New
356 Zealand^{69,127,139} (the Haast, Hokitika, Waiho and Whataroa Rivers); three draining the
357 Mackenzie Basin in Canada^{42,43,140-142} (the Peel, Arctic Red and Mackenzie Rivers); a
358 Taiwanese catchment^{41,43,59,86,93,143} (the Liwu River); and mountain catchments in
359 Guadeloupe^{144,145}. We can calculate the net rock-atmosphere CO₂ flux (J_{net} , tC km⁻² yr⁻¹) in
360 such regions¹⁴², with negative values representing a net atmospheric CO₂ sink, and positive
361 values a net CO₂ source:

$$362 \quad J_{net} = (-J_{OCbio-burial} + J_{OCpetro-ox}) + (-J_{sil-CO2} + J_{carb-sulf}) \quad (\text{Eq. 1}).$$

363 The first term represents the OC balance (OC_{biosphere} burial, $J_{OCbio-burial}$; and OC_{petro} oxidation,
364 $J_{OCpetro-ox}$), while the second term represents the inorganic components (silicate weathering
365 fluxes, $J_{sil-CO2}$; and CO₂ emissions from sulfide oxidation, $J_{carb-sulf}$). The assessments here are
366 presented with the caveat that modern-day fluxes determined from solutes ($J_{sil-CO2}$, $J_{carb-sulf}$,
367 $J_{OCpetro-ox}$) are directly compared to particulates ($J_{OCbio-burial}$), which have distinct sources of
368 uncertainty¹⁴⁶ and are potentially integrated over different time and spatial-scales^{28,147}.

369 Although most C cycle models have focused on parameterizing the rate of silicate
370 weathering^{24,75,148}, catchment-scale budgets highlight the importance of erosion-driven

371 $OC_{\text{biosphere}}$ fluxes as a CO_2 sink and erosion-driven sulfide weathering as a CO_2 source (Fig.
372 4). Notably, in the mountain catchments that are the most potent CO_2 sinks (the most
373 negative J_{net} values, such as for Whataroa), CO_2 draw down by erosion and burial of
374 $OC_{\text{biosphere}}$ is larger than that from by silicate weathering (Fig. 4a). The negative J_{net} values in
375 locations with large $J_{OC_{\text{bio-burial}}}$ terms emphasize that erosion can stimulate the organic
376 pathway of CO_2 drawdown and echo decades old interpretations from the Bengal Fan³⁷.

377 For catchments that are net sources of CO_2 during weathering and erosion (such as the
378 Liwu River), or within uncertainty of CO_2 neutral (such as the Arctic Red), $J_{\text{carb-sulf}}$ and
379 $J_{OC_{\text{petro-ox}}}$ are both important source terms (Fig. 4a). Consequently, exhumation and oxidation
380 of sulfides and OC_{petro} might be able to tip the net rock-atmosphere CO_2 balance of a
381 catchment from a sink to a source.

382 **4. Insights into processes and mechanisms**

383 When considered holistically, modern-day C fluxes in river catchments provide
384 critical information about how the geological C cycle operates (Fig. 3&4). However, C fluxes
385 are calculated from measurements made over short time windows, typically years to decades,
386 and can have large uncertainties (Fig. 4a). The C fluxes are also influenced by several factors
387 that can confound comparisons across spatial gradients in erosion rate, such as co-varying
388 changes in climate, lithology, and biology. Other observations and modelling efforts can
389 provide complementary process-level information. In this section, we discuss such
390 approaches, following them from studies of actively eroding hillslopes, across floodplains,
391 and into sedimentary archives that record past changes in erosion, weathering, and OC
392 transfers.

393 **4.1 A critical zone perspective**

394 Weathering profiles developed in rock and soil capture the sequence of chemical
395 reactions and physical transformations that occur as uplifting rock interacts with fluids near
396 the Earth's surface¹⁴⁹⁻¹⁵¹. Observations of the mineralogy, geochemistry, and physical
397 microstructure of these profiles can reveal mechanisms that link weathering and erosion, such
398 as porosity generation, mineral expansion, and micro-fracturing^{31,152-154}. Key weathering
399 processes, such as silicate mineral acid hydrolysis and sulfide oxidation, can extend to
400 considerable depths (10s of m) below the soil zone in the weathered bedrock^{151,155}. Fracturing
401 can also play a central role in weathering by facilitating water-rock interaction and the

402 penetration of O₂ and CO₂ to similar depths^{30,156,157}. The relationship between weathering and
403 fracturing is not fully understood, but might be critical to developing a mechanistic
404 understanding of how erosion drives weathering of silicates, sulfides, and OC_{petro}, and
405 associated C fluxes (ref.¹²³).

406 Models of weathering front propagation complement observations from weathering
407 profiles^{27,28,34,123,154,158}. Front propagation models are typically based on reactive transport
408 simulations of fluids percolating through the subsurface. Model results highlight how
409 weathering depends on mineral reactivity as well as fluid flow rates, both of which might
410 interact in important ways with erosional drivers^{34,158,159}. However, applying such models to
411 rapidly eroding terrains remains a challenge, owing to the incomplete understanding of fluid
412 flow and solute generation at catchment scales¹⁵⁹, as well as the geomorphic complexity of
413 actively eroding landscapes, where erosion and weathering rarely progress through simple
414 vertical profiles^{94,95,138}. Instead, stochastic erosion processes such as mass wasting, followed
415 by transport through sedimentary systems, might exert important but poorly understood
416 controls on weathering in these rapidly eroding landscapes.

417 **4.2 Landslide erosion and C transfers**

418 Landslides are one of the main erosional agents in mountains⁹⁴, and their role in the
419 transfer of OC_{biosphere} has been well characterized^{100,105,161-162}. In addition, landslide deposits
420 might be important as weathering reactors (with lithology-dependent implications on the C
421 cycle), owing to the chemical alteration of freshly mobilised rock in landslide sediment
422 deposits or hillslope scars¹⁶³⁻¹⁶⁵. The residence time of landslide material in the landscape sets
423 the timescale over which reactions take place¹⁶³ and thus influences the relative C fluxes from
424 phases which react quicker (sulfides, carbonates) and slower (OC_{petro}, silicate minerals)¹⁶⁴.
425 The residence time of landslide material is determined by the erosional processes that remove
426 landslide debris, such as fluvial transport¹⁶⁶ — suggesting that further study of landslide-
427 dominated settings could yield interesting process-level insights to complement critical zone
428 investigations.

429 One of the challenges in studying landslides is that they are stochastic and widely
430 distributed. Large populations of landslides triggered during major earthquakes provide an
431 opportunity to examine landslide C transfers, and the resulting erosional pulses could
432 contribute a substantial component of the denudation flux in many settings¹⁶⁷. Landslides

433 associated with the 2008 Mw 7.9 Wenchuan earthquake doubled the OC_{biosphere} supply to
434 rivers in the two years that followed the earthquake¹⁶⁸. In New Zealand lake records, ~40% of
435 the OC deposited over the last millennia was associated with the aftermath of four large
436 earthquakes¹⁶⁹. Earthquakes can also perturb solute fluxes, by chemical weathering of
437 landslide material or by changing subsurface hydrology¹⁷⁰. Therefore, active mountain belts
438 might influence C transfers via discrete, pulsed landslide erosion events, including those
439 associated with large earthquakes. Linking event-scale measurements with the implications
440 for the long-term C cycle remains an area for worthwhile study.

441 ***4.3 The role of floodplains***

442 Some mountainous rivers, such as those draining high-standing islands and active
443 margins, deliver material directly to the ocean, facilitating OC_{biosphere} burial and, in some
444 cases, reburial of eroded OC_{petro} (refs.^{59,130,171,172}). Other rivers feed large continental
445 floodplains that can serve as biogeochemical reactors^{65,173}. In such systems, OC_{biosphere} can be
446 stored over millennial timescales in floodplain, lake, and wetland sediments^{78,174,175},
447 facilitating the degradation of OC_{biosphere}. However, floodplains might also see the
448 replacement of mountain-derived OC with lowland-derived OC, as revealed by biomarkers
449 and their isotopes in the Amazon, Ganges and Congo systems¹⁷⁶⁻¹⁷⁹. OC replacement can
450 maintain, or indeed increase, OC_{biosphere} transfer to marine depocentres³⁹. Floodplain transit
451 can also be important for OC_{petro}, by providing additional opportunities for otherwise un-
452 weathered components to be oxidised during their journey^{119,120,180}.

453 The role of floodplains as inorganic weathering reactors is also increasing well-
454 understood. Eroded sulfide minerals are almost always weathered away in modern systems as
455 they are typically oxidized over transport lengths of only a few tens of km^{131,181}. Since silicate
456 minerals react more slowly, they are more likely to persist and be transported into and across
457 floodplains – making these settings potentially important for further reaction^{19,182}. The
458 Ganges floodplain contributes an important part of the silicate-derived alkalinity flux
459 delivered to the oceans by the Ganges River^{183,184}, as does the Madeira River floodplain of
460 the Amazon Basin^{132,185,186}. Despite the recognition that floodplains have a vital role to play
461 in geochemical carbon transfers, quantification of the associated net rock-atmosphere
462 exchange remains out of reach.

463 ***4.4 Records from geological archives***

464 Sediments can preserve a direct record of the OC transferred to the rock reservoir, as
465 well as the chemical signature of rock weathering (reflected in the mineralogy and elemental
466 composition of the sediments), thus providing insights into key relationships in the erosion-C
467 cycle system. However, it is often challenging to extract chemical signatures of weathering
468 from sedimentary archives because imbalances in the C cycle of ~1-5% can have large effects
469 over million-year timescales¹⁷, yet geological records may not record past changes with such
470 fidelity. Nevertheless, detrital records from around the Asian margin have been interpreted to
471 reflect the weathering response to the interaction of mountain uplift and climatic changes
472 over timescales ranging from the Last Glacial Maximum^{187,188} to the millions of years
473 associated with tectonic uplift and evolution of the Asian monsoon^{189,190}. Elsewhere, detrital
474 records¹⁹¹ have also been used to reconstruct changes in weathering intensity and, when
475 paired with data on paleo-denudation rates, to infer the coupling between erosion and
476 weathering over glacial-interglacial timescales¹⁹².

477 Seawater chemistry records, including radiogenic strontium (Sr) and osmium (Os)
478 isotopes,^{11,193} track changes in continental weathering intensity and/or fluxes¹⁹⁴. Such isotopic
479 records can shed light on how erosion might force the C cycle. For instance, over the past 50
480 Ma, the combination of the Os and Sr isotopes reveal a shift in the balance of sulfide
481 oxidation to silicate weathering, highlighting an increase in the net draw down of CO₂ by
482 mountain uplift between ~10-30 Ma (ref.⁴³). Another example is recent work combining
483 Neogene lithium isotope¹⁹⁵ and beryllium isotope¹⁹⁶ records, together with reconstructions of
484 atmospheric CO₂ concentrations¹⁹⁷, to suggest that a doubling of global erosion rates over the
485 last 12 Ma changed the climate sensitivity to silicate weathering fluxes, thus moderating
486 atmospheric CO₂ (ref.⁷⁵).

487 The stable isotopes of carbon ($\delta^{13}\text{C}$), recorded both in marine carbonate minerals and
488 OC from sedimentary rocks, have been used to quantify past fluxes of carbon at the Earth's
489 surface and between the major sedimentary reservoirs^{36,81,194,198,199}. The $\delta^{13}\text{C}$ of carbonate
490 rocks and organic material encode relative changes in the total OC burial flux compared to
491 the fluxes from weathering and carbonate burial (Fig. 1b). Isotope mass balance models
492 based on $\delta^{13}\text{C}$ variations over the last 60 million years suggest that OC burial consistently
493 represented 25±6% of the total C removal from the ocean and atmosphere system⁸¹, which
494 suggests that C removal by OC burial and carbonate burial generally increased and decreased
495 in tandem. The stability in this ratio of C burial implies a somewhat remarkable coupling,

496 whereby the weathering supply of Ca and Mg to the oceans is mirrored in the burial and
497 preservation of OC. The positive relationship between OC_{biosphere} erosion and physical erosion
498 (Figs. 3b) might explain this relationship, as erosion-driven increases (or decreases) in silicate
499 weathering might be accompanied by changes in erosion and transfer of OC_{biosphere}. The
500 enhanced supply of rock-derived nutrients to the ocean, as a result of higher weathering
501 fluxes, might also feed marine productivity and OC burial. However, caution is warranted
502 when interpreting C isotope records, in part because they are sensitive to changes in the
503 isotopic composition of CO₂ that is degassed from the solid Earth^{199,200}.

504 **5. A new view of erosion and C transfers**

505 With new information emerging regarding C sources and sinks in the present day, as well as
506 in the geological past, we are poised to consider how recent insights influence our view of the
507 long-term C cycle — and how human-induced changes in erosions rates might influence the
508 global C cycle moving forward.

509 ***5.1 A long-term C cycle view***

510 In this Review we have made the case that it is essential to take a holistic view of all
511 CO₂ sources and sinks associated with uplift and erosion to fully evaluate the long-standing
512 hypotheses that link mountain building to the evolution of global climate. Our Review reveals
513 that there is considerable variability in the net C budget of erosion (Figs. 3, 4), indicating that
514 the total CO₂ budget might depend on multiple factors.

515 Lithology matters (Fig. 4), both in terms of the reactivity of silicate minerals and the
516 abundance of sulfide and carbonate phases. In this respect, volcanic rocks are optimized for
517 CO₂ drawdown (Fig. 5), with abundant cation-rich primary minerals that can sustain high
518 silicate weathering rates²⁰¹ and few sulfide minerals or OC_{petro}. Young, rapidly eroding
519 volcanic terrains^{144,202,203} are thus poised to play important roles in climate evolution²⁰⁴.
520 Sedimentary rocks, in contrast, are typically depleted in base cations due to past weathering
521 cycles¹⁴⁷, and shales can be rich in sulfides and OC_{petro}. As a result, they are primed to be
522 potent CO₂ sources^{41,42,124,134} (Fig. 3b). Thus, the net C cycle fluxes associated with mountain
523 building might vary depending on regional tectonics, and perhaps throughout orogenic
524 evolution^{2,205} as different lithologies are exposed (for example, initial erosion of arc volcanics
525 and subsequent erosion of a sedimentary pile as exhumation continues).

526 In addition, the temporal sequence of mountain building might influence the long-
527 term C cycle because denudation rates evolve over time. Our Review indicates that there
528 could be a “sweet spot” of net CO₂ drawdown at intermediate denudation rates. At the lowest
529 denudation rates, silicate weathering is supply limited and OC_{biosphere} erosion muted⁵⁶.
530 However, at the highest denudation rates, oxidative weathering surpasses silicate weathering,
531 and the net balance tips towards a CO₂ source, at least for some lithologies. Therefore, during
532 periods of active uplift (such as in Taiwan today), mountain building might push the system
533 towards becoming a CO₂ source^{41,134,206} (depending on the sink through OC_{biosphere}
534 burial^{59,127}). In the later stages of evolution (for example, the modern relicts of the
535 Appalachian orogeny) CO₂ sinks can dominate, though the overall fluxes might be lower
536 (Figs. 4b, 5). While we can predict these general trends, complexities arising from the
537 interplay of erosion, weathering and climate preclude the definition of specific thresholds for
538 the transition from a net source to a net sink of CO₂ in mountainous regions, presenting
539 challenges for quantitative modelling (Fig. 3).

540 Climate might also modulate the effect of erosion on the C cycle in complex ways
541 (Fig. 5). Cooler temperatures and lower runoff decrease silicate weathering yields^{32,207} and
542 reduce erosion and transfer of OC_{biosphere} (refs.^{92,104}). At the same time, glaciation could
543 increase CO₂ emissions via oxidative weathering in basins dominated by sedimentary
544 rocks^{62,127} through a combination of physical (frost shattering and surface area production)
545 and biogeochemical (higher relative O₂ availability for oxidative weathering) mechanisms.
546 Although the lithological control must still be considered, erosion in warmer, wetter climates
547 might be more likely to produce a net sink of CO₂ than erosion in colder climates, at least
548 where frost-cracking and glacial processes become active (Fig. 5). Thus, the location of
549 eroding mountainous topography with respect to latitude, and the timing of mountain uplift
550 with respect to climate evolution, likely play important roles in determining the overall effect
551 of erosion on the global C cycle.

552 The impact of erosion on the C cycle might have changed over geological time as the
553 composition of exposed rocks and the atmosphere evolved. For example, the O₂-rich
554 atmosphere, terrestrial ecosystems and large sedimentary stocks of carbonate and OC_{petro} that
555 characterise the mid- to late-Phanerozoic could have aided the oxidative reactions that drive
556 CO₂ emissions. Conversely, heightened erosion and burial of OC_{biosphere} in the Phanerozoic
557 might have countered the increased CO₂ emissions. By contrast, mountain building events

558 earlier in Earth's history, such as during the formation of the supercontinent Rodinia, 1100-
559 900 Ma ago, would likely have had very different C cycle impacts^{64,81}.

560 **5.2 An Anthropocene view**

561 Variations in erosional carbon fluxes might influence the global carbon cycle over
562 much shorter timescales, such as over hundreds of years. Over the coming century, human-
563 induced changes in erosion-related CO₂ emissions from sedimentary rocks could result in a
564 previously overlooked source of C to the atmosphere. Humans have perturbed erosion rates
565 and global sediment fluxes²⁰⁸, causing erosion rates in some mountain catchments underlain
566 by shales to more than double during the industrial period^{209,210}. There is also evidence that
567 the legacy of coal mining can vastly increase oxidative weathering reactions, releasing CO₂
568 from sulfuric acid-carbonate weathering and OC_{petro} oxidation²¹¹. An average increase in
569 global erosion of ~10-20% is likely to correspond to a similar increase in CO₂ emissions from
570 weathering of shales (Fig. 3c&d), potentially prolonging the impacts of anthropogenic
571 climate change. Thus, the assumptions that carbonate weathering is CO₂ neutral in terms of
572 the human-modified carbon cycle²¹², and that OC_{petro} is unreactive and passive in soils⁸² (and
573 thus not a CO₂ source), must be revisited.

574 Erosive environments not directly altered by humans might also respond to
575 anthropogenic climate change. At high latitudes, thermokarst erosion in northern Canada
576 associated with permafrost thaw exposes carbonate and sulfide minerals in scars and deposits.
577 The resulting sulfuric acid-carbonate weathering could release much more CO₂ than the
578 degradation of thawed OC in permafrost²¹³. Changes in the cryosphere could also influence
579 the balance between CO₂ sources and sinks, with glacier mass loss exposing glaciogenic
580 sediments to oxidative weathering^{62,127}. The fluxes associated with each process are
581 uncertain, but they could contribute to regional scale carbon budgets, and potentially offset
582 part of the net land sink of anthropogenic CO₂ (ref.⁴⁸). Understanding how human-induced
583 changes in land use, erosion, temperature and runoff control the CO₂ emissions from
584 weathering sedimentary rocks^{41,62,127,213} will be important for evaluating their potential role as
585 CO₂ sources that extend the impacts of climate change²¹⁴.

586 In considering the effect of human induced erosion, it is important to note that the
587 combustion of fossil fuels itself represents accelerated oxidation of OC_{petro} — and the
588 magnitude of this flux is very large. The 'natural' flux of CO₂ released by OC_{petro} oxidation⁴⁰

589 is 40-100 MtC yr⁻¹, while the release of OC by fossil fuel burning²¹⁵ is 921400 MtC yr⁻¹ (Fig.
590 2). Similarly, the combined CO₂ sink by OC burial and silicate weathering (260-310 MtC yr⁻¹;
591 Fig. 1B) is only ~20% of the estimated CO₂ release associated with deforestation and land
592 use change (~1500 MtC yr⁻¹). As such, doubling the global CO₂ release from erosion-driven
593 oxidative weathering of OC_{petro} and sulfides (to ~280 MtC yr⁻¹) would only equate to ~3% of
594 anthropogenic CO₂ emissions. Even so, the long-term effects of human-heightened erosion
595 could be important and, over timescales of >1000 years, could influence the re-distribution of
596 carbon from human CO₂ emissions via silicate weathering and OC burial (Fig. 2), thus
597 shaping the future trajectory of atmospheric CO₂ concentrations²¹⁶.

598 **6 Conclusions**

599 **6.1 Summary**

600 Multiple processes determine the net “rock-atmosphere” exchange of C. CO₂ is drawn
601 down by silicate weathering and OC_{biosphere} erosion and burial, but it is also released by
602 OC_{petro} oxidation and sulfuric acid weathering. Understanding the overall effect of erosion
603 and mountain building on the C cycle depends on considering all C transfer fluxes together.
604 In many present-day systems (Fig. 4), the C balance is strongly influenced by fluxes that are
605 overlooked in existing geochemical models of the long term C cycle: sulfide oxidation,
606 OC_{biosphere} erosion and burial, and OC_{petro} oxidation.

607 The fluxes associated with the C transfer mechanisms that shape the geological C
608 cycle all broadly increase with erosion rate (Fig. 3); erosion removes OC from vegetation and
609 soils and enhances the supply of minerals to the critical zone, where water, acids and rocks
610 interact. However, variability exists in the relationships between erosion rate and the flux of
611 C from each transfer mechanism, which could, in part, reflect climatic (hydrological and
612 temperature) controls. In terms of CO₂ sinks, silicate weathering reactions are most sensitive
613 to changes in hydrology and temperature in mountainous regions. In addition, erosion of
614 OC_{biosphere} is also linked to changes in runoff and, therefore, the CO₂ sinks can drive negative
615 feedbacks in the carbon cycle — feedbacks that are sustained by erosion. CO₂ sources via
616 oxidative weathering are typically considered to be only weakly dependent on climate,
617 although there is some evidence to suggest that a stronger relationship might exist for the
618 oxidation of OC_{petro}. CO₂ sources might also be sensitive to climate indirectly through
619 facilitation of oxidative weathering by glacial processes.

620 Lithology appears to play a central role in determining the balance between the
621 different C fluxes. A mountain range underlain by sedimentary-rocks might produce a net
622 CO₂ source to the atmosphere (or be CO₂ neutral), whereas volcanic rocks are more likely to
623 be efficient CO₂ sinks. Understanding the impact of mountain building on the C cycle over
624 geologic time will depend on quantifying the interaction of lithologic, climatic, and erosional
625 controls (Fig. 5). Such understanding would benefit from future catchment-based estimates of
626 the net “rock-atmosphere” CO₂ transfers (Fig. 4) across a wider range of scales, lithologies,
627 and climate conditions, and will require C cycle models that can tackle the four key processes
628 that are operating as CO₂ sources and sinks.

629 **6.2 Future perspectives**

630 We suggest that future research should focus on several areas that will help determine
631 the relationship between mountain building, heightened erosion and CO₂ sources and sinks.
632 One set of future directions relates to the fluxes and controls on oxidative weathering and C
633 release. More work is required to understand the mechanism of sulfide and OC_{petro} oxidation,
634 and to quantify their associated CO₂ release. Future research will benefit from studies at a
635 wide range of scales, including measurements at the weathering profile scale (over several
636 meters) of specific rock-types to investigate lithology-dependence, as well as assessment of
637 net CO₂ fluxes from large river basins to constrain landscape-scale controls. In addition,
638 future studies should aim to understand the role of climatic factors, such as temperature and
639 hydrology, on the CO₂ release by weathering of shales and sedimentary rocks through OC_{petro}
640 and sulfide oxidation. A combination of field, laboratory and numerical modelling
641 approaches will be needed to understand potential feedbacks between climate and CO₂
642 emissions by oxidative weathering.

643 The role of floodplains in the net CO₂ transfer between rocks and the atmosphere also
644 needs to be addressed, as do chemical reactions within landslide deposits that potentially act
645 as weathering hotspots in actively eroding landscapes. Better understanding of where
646 weathering takes place within landscapes could help to resolve discrepancies between soil
647 and solute-derived silicate weathering (Fig. 3a). Furthermore, the role of weathering and
648 erosion in supplying rock-derived nutrients to terrestrial and marine ecosystems is
649 increasingly recognized, but more work is needed to quantify the net impact on long-term C
650 storage.

651 Future analyses of catchment-scale C budgets, to assess the net rock-atmosphere CO₂
652 exchange in different settings, are also encouraged. Current data only allow for the full C
653 budget to be assessed in one volcanic setting (Fig. 4), even though volcanic rocks are
654 recognised as key locations for silicate weathering and CO₂ drawdown. Such studies would
655 provide the empirical basis for models that consider all C transfer mechanisms holistically,
656 and thus help to understand how mountain building and erosion impacts global
657 biogeochemical cycles. There is now an opportunity to design and test C cycle models that
658 include the erosion and burial of OC_{biosphere} and the oxidative weathering of OC_{petro} and
659 sulfide minerals and, therefore, shift the focus beyond the canonical focus on silicate
660 weathering.

661 7. References:

- 662 1. Ahnert, F. (1970) Functional relationships between denudation, relief, and uplift in
663 large, mid-latitude drainage basins, *American Journal of Science*, 268, 243-263.
664 <https://doi.org/10.2475/ajs.268.3.243>
- 665 2. Dewey, J. F., & Horsfield, B. (1970). Plate tectonics, orogeny and continental growth.
666 *Nature*. <https://doi.org/10.1038/225521a0>
- 667 3. Hager, B. H., & Richards, M. A. (1989). Long-Wavelength Variations in Earth's
668 Geoid: Physical Models and Dynamical Implications. *Philosophical Transactions of the*
669 *Royal Society A: Mathematical, Physical and Engineering Sciences*, 328(1599), 309–
670 327. <https://doi.org/10.1098/rsta.1989.0038>
- 671 4. Willett, S. D. (1999). Orogeny and orography: The effects of erosion on the structure of
672 mountain belts. *Journal of Geophysical Research: Solid Earth*, 104(B12), 28957–
673 28981. <https://doi.org/10.1029/1999jb900248>
- 674 5. Braun, J. (2010). The many surface expressions of mantle dynamics. *Nature*
675 *Geoscience*, 3(12), 825–833. <https://doi.org/10.1038/ngeo1020>
- 676 6. Milliman, J. D., & Syvitski, J. P. M. (1992). Geomorphic/tectonic control of sediment
677 discharge to the ocean: the importance of small mountainous rivers. *Journal of*
678 *Geology*, 100(5), 525–544. <https://doi.org/10.1086/629606>
- 679 7. Métivier, F., Gaudemer, Y., Tapponnier, P., & Klein, M. (1999). Mass accumulation
680 rates in Asia during the Cenozoic. *Geophysical Journal International*.
681 <https://doi.org/10.1046/j.1365-246X.1999.00802.x>
- 682 8. **Chamberlin, T. C. (1899). An Attempt to Frame a Working Hypothesis of the**
683 **Cause of Glacial Periods on an Atmospheric Basis. *The Journal of Geology*.**
684 **<https://doi.org/10.1086/608524> — First paper (as far as known) to propose mountain**
685 **building as a driver of atmospheric CO₂ drawdown and global cooling, suggesting**
686 **this mechanism as the most plausible explanation for episodes of glaciation in the**
687 **geologic past.**
- 688 9. Richter, F. M., Rowley, D. B., & DePaolo, D. J. (1992). Sr isotope evolution of
689 seawater: the role of tectonics. *Earth and Planetary Science Letters*.
690 [https://doi.org/10.1016/0012-821X\(92\)90070-C](https://doi.org/10.1016/0012-821X(92)90070-C)

- 691 10. Molnar, P. (1988). A review of geophysical constraints on the deep structure of the
692 Tibetan Plateau, the Himalaya and the Karakoram, and their tectonic implications.
693 *Philosophical Transactions - Royal Society of London, Series A*, 326(1589), 33–88.
694 <https://doi.org/10.1098/rsta.1988.0080>
- 695 11. Miller, K. G., Fairbanks, R. G., & Mountain, G. S. (1987). Tertiary oxygen isotope
696 synthesis, sea level history, and continental margin erosion. *Paleoceanography*, 2(1),
697 1–19. <https://doi.org/10.1029/PA002i001p00001>
- 698 12. **Raymo, M. E., Ruddiman, W. F., & Froelich, P. N. (1988). Influence of late
699 Cenozoic mountain building on ocean geochemical cycles. *Geology*.
700 [https://doi.org/10.1130/0091-7613\(1988\)016<0649:IOLCMB>2.3.CO;2](https://doi.org/10.1130/0091-7613(1988)016<0649:IOLCMB>2.3.CO;2) —
701 *Pioneering study connecting evolution of Tibetan Plateau uplift, changes in marine
702 chemistry, and global cooling over the past 60 Myrs; stimulated a Renaissance in
703 efforts to understanding links between mountain building and climate.***
- 704 13. Raymo, M. E., & Ruddiman, W. F. (1992). Tectonic forcing of late Cenozoic climate.
705 *Nature*. <https://doi.org/10.1038/359117a0>
- 706 14. Volk, T. (1993). Cooling in the late Cenozoic [12]. *Nature*.
707 <https://doi.org/10.1038/361123a0>
- 708 15. Caldeira, K., Arthur, M. A., Berner, R. A., & Lasaga, A. C. (1993). Cooling in the late
709 Cenozoic [13]. *Nature*, Vol. 361, pp. 123–124. <https://doi.org/10.1038/361123b0>
- 710 16. Raymo, M. E., & Ruddiman, W. (1993). Cooling in the late Cenozoic [14]. *Nature*.
711 <https://doi.org/10.1038/361124a0>
- 712 17. Berner, R. A., & Caldeira, K. (1997). The need for mass balance and feedback in the
713 geochemical carbon cycle. *Geology*. [https://doi.org/10.1130/0091-
714 7613\(1997\)025<0955:TNFMBA>2.3.CO;2](https://doi.org/10.1130/0091-7613(1997)025<0955:TNFMBA>2.3.CO;2)
- 715 18. **Gaillardet, J., Dupré, B., Louvat, P., & Allègre, C. J. (1999). Global silicate
716 weathering and CO₂ consumption rates deduced from the chemistry of large
717 rivers. *Chemical Geology*. [https://doi.org/10.1016/S0009-2541\(99\)00031-5](https://doi.org/10.1016/S0009-2541(99)00031-5) —
718 *Assembled and analyzed a river chemistry database to provide landmark estimates of
719 silicate weathering fluxes and their global controls, revealing broad relationship
720 between atmospheric CO₂ drawdown by weathering and erosion rates.***
- 721 19. Galy, A., & France-Lanord, C. (1999). Weathering processes in the Ganges-
722 Brahmaputra basin and the riverine alkalinity budget. *Chemical Geology*.
723 [https://doi.org/10.1016/S0009-2541\(99\)00033-9](https://doi.org/10.1016/S0009-2541(99)00033-9)
- 724 20. White, A. F., & Brantley, S. L. (2003). The effect of time on the weathering of silicate
725 minerals: Why do weathering rates differ in the laboratory and field? *Chemical
726 Geology*. <https://doi.org/10.1016/j.chemgeo.2003.03.001>
- 727 21. Ebelmen, J. (1845). Sur les produits de la décomposition des especes minérales de
728 famille des silicates, *Ann. Mines*, 7 (1845), 3–66.
- 729 22. Urey, H. C. (1952). On the Early Chemical History of the Earth and the Origin of Life.
730 *Proceedings of the National Academy of Sciences*.
731 <https://doi.org/10.1073/pnas.38.4.351>
- 732 23. Walker, J. C. G., Hays, P. B., & Kasting, J. F. (1981). A negative feedback mechanism
733 for the long-term stabilization of Earth's surface temperature. *Journal of Geophysical
734 Research*. <https://doi.org/10.1029/JC086iC10p09776>

- 735 24. Berner, R. A., Lasaga, A. C., & Garrels, R. M. (1983). The carbonate-silicate
736 geochemical cycle and its effect on atmospheric carbon dioxide over the past 100
737 million years. *American Journal of Science*. <https://doi.org/10.2475/ajs.283.7.641>
- 738 25. Anderson, S. P. (2019). Breaking it Down: Mechanical Processes in the Weathering
739 Engine. *Elements*, 15(4), 247–252. <https://doi.org/10.2138/gselements.15.4.247>
- 740 26. Lasaga, A. C., Soler, J. M., Ganor, J., Burch, T. E., & Nagy, K. L. (1994). Chemical
741 weathering rate laws and global geochemical cycles. *Geochimica et Cosmochimica*
742 *Acta*. [https://doi.org/10.1016/0016-7037\(94\)90016-7](https://doi.org/10.1016/0016-7037(94)90016-7)
- 743 27. Maher, K. (2010). The dependence of chemical weathering rates on fluid residence
744 time. *Earth and Planetary Science Letters*. <https://doi.org/10.1016/j.epsl.2010.03.010>
- 745 28. **Maher, K. (2011). The role of fluid residence time and topographic scales in**
746 **determining chemical fluxes from landscapes. *Earth and Planetary Science Letters*.**
747 **<https://doi.org/10.1016/j.epsl.2011.09.040> — *Developed a mechanistic framework***
748 ***for understanding why hydrology is a primary control on weathering fluxes,***
749 ***revealing the key role of saturation state in influencing reaction rates in natural***
750 ***weathering systems.***
- 751 29. Brantley, S. L., & Lebedeva, M. (2011). Learning to Read the Chemistry of Regolith to
752 Understand the Critical Zone. *Annual Review of Earth and Planetary Sciences*.
753 <https://doi.org/10.1146/annurev-earth-040809-152321>
- 754 30. Clair, J. S., Moon, S., Holbrook, W. S., Perron, J. T., Riebe, C. S., Martel, S. J., ... De
755 Richter, D. B. (2015). Geophysical imaging reveals topographic stress control of
756 bedrock weathering. *Science*. <https://doi.org/10.1126/science.aab2210>
- 757 31. **Gu, X., Rempe, D. M., Dietrich, W. E., West, A. J., Lin, T. C., Jin, L., & Brantley,**
758 **S. L. (2020). Chemical reactions, porosity, and microfracturing in shale during**
759 **weathering: The effect of erosion rate. *Geochimica et Cosmochimica Acta*.**
760 **<https://doi.org/10.1016/j.gca.2019.09.044> — *Revealed how rock structure varies***
761 ***across erosion rates in ways that may mechanistically explain how erosion and***
762 ***weathering are linked.***
- 763 32. **West, A. J., Galy, A., & Bickle, M. (2005). Tectonic and climatic controls on**
764 **silicate weathering. *Earth and Planetary Science Letters*.**
765 **<https://doi.org/10.1016/j.epsl.2005.03.020> — *Revealed the supply vs kinetic***
766 ***limitation of silicate weathering across river catchments, as a function of erosion***
767 ***rates. By linking the empirical data to a predictive model, the role of erosion rate,***
768 ***temperature and runoff could be de-convolved for the first time.***
- 769 33. Gabet, E. J., & Mudd, S. M. (2009). A theoretical model coupling chemical weathering
770 rates with denudation rates. *Geology*. <https://doi.org/10.1130/G25270A.1>
- 771 34. Maher, K., & Chamberlain, C. P. (2014). Hydrologic regulation of chemical weathering
772 and the geologic. *Science*. <https://doi.org/10.1126/science.1250770>
- 773 35. Gaillardet, J., & Galy, A. (2008). Himalaya-carbon sink or source? *Science*, 320, 1727-
774 1728. <https://doi.org/10.1126/science.1159279>
- 775 36. Derry, L. A., & France-Lanord, C. (1996). Neogene Himalayan weathering history and
776 river ⁸⁷Sr/⁸⁶Sr: Impact on the marine Sr record. *Earth and Planetary Science Letters*.
777 [https://doi.org/10.1016/0012-821x\(96\)00091-x](https://doi.org/10.1016/0012-821x(96)00091-x)
- 778 37. **France-Lanord, C., & Derry, L. A. (1997). Organic carbon burial forcing of the**
779 **carbon cycle from Himalayan erosion. *Nature*. <https://doi.org/10.1038/36324> —**
780 ***Discovered very high fluxes of organic carbon burial in the Bengal Fan as a result of***

- 781 ***Himalayan erosion, far out-pacing silicate weathering in this system and giving birth***
782 ***to new lines of research to understand erosional controls on the organic carbon***
783 ***cycle.***
- 784 38. Burdige, D. J. (2005). Burial of terrestrial organic matter in marine sediments: A re-
785 assessment. *Global Biogeochemical Cycles*. <https://doi.org/10.1029/2004GB002368>
- 786 39. Galy, V., France-Lanord, C., Beyssac, O., Faure, P., Kudrass, H., & Palhol, F. (2007).
787 Efficient organic carbon burial in the Bengal fan sustained by the Himalayan erosional
788 system. *Nature*, 450(7168), 407–410. <https://doi.org/10.1038/nature06273>
- 789 40. Petsch, S. T. (2013). Weathering of Organic Carbon. In *Treatise on Geochemistry: Second Edition*.
790 <https://doi.org/10.1016/B978-0-08-095975-7.01013-5>
- 791 41. Hilton, R. G., Gaillardet, J. Ô., Calmels, D., & Birck, J. L. (2014). Geological
792 respiration of a mountain belt revealed by the trace element rhenium. *Earth and*
793 *Planetary Science Letters*, 403, 27–36. <https://doi.org/10.1016/j.epsl.2014.06.021> —
794 ***Developed a new proxy that enables measurement of petrogenic carbon oxidation***
795 ***fluxes at the scale of river catchments, and found a link between oxidation fluxes and***
796 ***physical erosion rate.***
- 797 42. Calmels, D., Gaillardet, J., Brenot, A., & France-Lanord, C. (2007). Sustained
798 sulfide oxidation by physical erosion processes in the Mackenzie River basin:
799 Climatic perspectives. *Geology*. <https://doi.org/10.1130/G24132A.1> — ***Revealed***
800 ***correlation between erosion rate and fluxes from sulfate oxidation, opening up this***
801 ***field of inquiry.***
- 802 43. Torres, M. A., West, A. J., & Li, G. (2014). Sulphide oxidation and carbonate
803 dissolution as a source of CO₂ over geological timescales. *Nature*.
804 <https://doi.org/10.1038/nature13030> — ***Proposed that sulfide oxidation can have***
805 ***important effects on the carbon cycle over long periods of time (up to 10s of Myrs),***
806 ***demonstrated links with erosion, and explored importance over the past 60 Myrs.***
- 807 44. Plank, T., & Manning, C. E. (2019). Subducting carbon. *Nature*, Vol. 574, pp. 343–
808 352. <https://doi.org/10.1038/s41586-019-1643-z>
- 809 45. Wong, K., Mason, E., Brune, S., East, M., Edmonds, M., & Zahirovic, S. (2019). Deep
810 Carbon Cycling Over the Past 200 Million Years: A Review of Fluxes in Different
811 Tectonic Settings. *Frontiers in Earth Science*, 7.
812 <https://doi.org/10.3389/feart.2019.00263>
- 813 46. Kerrick, D. M., & Caldeira, K. (1998). Metamorphic CO₂ degassing from orogenic
814 belts. *Chemical Geology*. [https://doi.org/10.1016/S0009-2541\(97\)00144-7](https://doi.org/10.1016/S0009-2541(97)00144-7)
- 815 47. Becker, J. A., Bickle, M. J., Galy, A., & Holland, T. J. B. (2008). Himalayan
816 metamorphic CO₂ fluxes: Quantitative constraints from hydrothermal springs. *Earth*
817 *and Planetary Science Letters*. <https://doi.org/10.1016/j.epsl.2007.10.046>
- 818 48. Ciais, P., C. Sabine, G. Bala, L. Bopp, V. Brovkin, J. Canadell, A. Chhabra, R. DeFries,
819 J. Galloway, M. Heimann, C. Jones, C. Le Quéré, R.B. Myneni, S. Piao and P.
820 Thornton, 2013: Carbon and Other Biogeochemical Cycles. In: *Climate Change 2013:*
821 *The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment*
822 *Report of the Intergovernmental Panel on Climate Change* [Stocker, T.F., D. Qin, G.-
823 K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M.
824 Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New
825 York, NY, USA.

- 826 49. Sundquist, E. T., & Visser, K. (2003). The Geologic History of the Carbon Cycle. In
827 *Treatise on Geochemistry*. <https://doi.org/10.1016/B0-08-043751-6/08133-0>
- 828 50. Holland, H. D., Lazar, B., & McCaffrey, M. (1986). Evolution of the atmosphere and
829 oceans. *Nature*. <https://doi.org/10.1038/320027a0>
- 830 51. Moon, S., Chamberlain, C. P., & Hilley, G. E. (2014). New estimates of silicate
831 weathering rates and their uncertainties in global rivers. *Geochimica et Cosmochimica*
832 *Acta*. <https://doi.org/10.1016/j.gca.2014.02.033>
- 833 52. Kump, L. R., & Arthur, M. A. (1997). Global Chemical Erosion during the Cenozoic:
834 Weatherability Balances the Budgets. In *Tectonic Uplift and Climate Change*.
835 https://doi.org/10.1007/978-1-4615-5935-1_18
- 836 53. Caves, J. K., Jost, A. B., Lau, K. V., & Maher, K. (2016). Cenozoic carbon cycle
837 imbalances and a variable weathering feedback. *Earth and Planetary Science Letters*.
838 <https://doi.org/10.1016/j.epsl.2016.06.035>
- 839 54. Berner, R. A., & Kothavala, Z. (2001). Geocarb III: A Revised Model of Atmospheric
840 CO₂ over Phanerozoic Time. *American Journal of Science*. 301, 182-204.
841 <https://doi.org/10.2475/ajs.301.2.182>
- 842 55. Bergman, N. M., Lenton, T. M., & Watson, A. J. (2004). COPSE: A new model of
843 biogeochemical cycling over Phanerozoic time. *American Journal of Science*. 304, 397-
844 437. <https://doi.org/10.2475/ajs.304.5.397>
- 845 56. Edmond, J. M. & Huh, Y. (2003). Non-steady state carbonate recycling and
846 implications for the evolution of atmospheric PCO₂. *Earth and Planetary Science*
847 *Letters*. 216, 125-139. [https://doi.org/10.1016/S0012-821X\(03\)00510-7](https://doi.org/10.1016/S0012-821X(03)00510-7)
- 848 57. Kump, L. R. (2018). Prolonged Late Permian–Early Triassic hyperthermal: failure of
849 climate regulation? *Phil. Trans. R. Soc. A*. 37620170078.
850 <http://doi.org/10.1098/rsta.2017.0078>
- 851 58. Isson, T. T., Planavsky, N. J., Coogan, L. A., Stewart, E. M., Ague, J. J., Bolton, E. W.,
852 et al. (2020). Evolution of the global carbon cycle and climate regulation on earth.
853 *Global Biogeochemical Cycles*, 34, e2018GB006061.
854 <https://doi.org/10.1029/2018GB006061>
- 855 59. Kao, S. J., Hilton, R. G., Selvaraj, K., Dai, M., Zehetner, F., Huang, J. C., ... Hovius,
856 N. (2014). Preservation of terrestrial organic carbon in marine sediments offshore
857 Taiwan: Mountain building and atmospheric carbon dioxide sequestration. *Earth*
858 *Surface Dynamics*, 2(1), 127–139. <https://doi.org/10.5194/esurf-2-127-2014>
- 859 60. Berner, R. A., & Canfield, D. E. (1989). A new model for atmospheric oxygen over
860 Phanerozoic time. *American Journal of Science*, 289(4), 333–361.
861 <https://doi.org/10.2475/ajs.289.4.333>
- 862 61. Hayes, J. M., & Waldbauer, J. R. (2006). The carbon cycle and associated redox
863 processes through time. *Philosophical Transactions of the Royal Society B: Biological*
864 *Sciences*. <https://doi.org/10.1098/rstb.2006.1840>
- 865 62. Torres, M. A., Moosdorf, N., Hartmann, J., Adkins, J. F., & West, A. J. (2017). Glacial
866 weathering, sulfide oxidation, and global carbon cycle feedbacks. *Proceedings of the*
867 *National Academy of Sciences of the United States of America*.
868 <https://doi.org/10.1073/pnas.1702953114>
- 869 63. Stolper, D. A., Bender, M. L., Dreyfus, G. B., Yan, Y., & Higgins, J. A. (2016). A
870 Pleistocene ice core record of atmospheric O₂ concentrations. *Science*.
871 <https://doi.org/10.1126/science.aaf5445>

- 872 64. Laakso, T. A., & Schrag, D. P. (2014). Regulation of atmospheric oxygen during the
873 Proterozoic. *Earth and Planetary Science Letters*. 388, 81-91.
874 <https://doi.org/10.1016/j.epsl.2013.11.049>
- 875 65. Mayorga, E., Aufdenkampe, A. K., Masiello, C. A., Krusche, A. V., Hedges, J. I.,
876 Quay, P. D., ... Brown, T. A. (2005). Young organic matter as a source of carbon
877 dioxide outgassing from Amazonian rivers. *Nature*, 436(7050), 538–541.
878 <https://doi.org/10.1038/nature03880>
- 879 66. Marx, A., Dusek, J., Jankovec, J., Sanda, M., Vogel, T., van Geldern, R., Hartmann, J.,
880 and Barth, J. A. C. (2017), A review of CO₂ and associated carbon dynamics in
881 headwater streams: A global perspective, *Rev. Geophys.*, 55, 560– 585,
882 doi:10.1002/2016RG000547.
- 883 67. Mackenzie, F. T., & Garrels, R. M. (1966). Chemical mass balance between rivers and
884 oceans. *American Journal of Science*. <https://doi.org/10.2475/ajs.264.7.507>
- 885 68. Larsen, I. J., Montgomery, D. R., & Greenberg, H. M. (2014). The contribution of
886 mountains to global denudation. *Geology*, 42(6), 527–530.
887 <https://doi.org/10.1130/G35136.1>
- 888 **69. Jacobson, A. D., & Blum, J. D. (2003). Relationship between mechanical erosion**
889 **and atmospheric CO₂ consumption in the New Zealand Southern Alps. *Geology*.**
890 **<https://doi.org/10.1130/G19662.1> — Brought to light the importance of**
891 ***distinguishing carbonate vs. silicate weathering in evaluating CO₂ drawdown,***
892 ***demonstrating that high erosion rates tend to produce increased proportion of solutes***
893 ***from carbonate sources.***
- 894 70. Stallard, R. F., & Edmond, J. M. (1983). Geochemistry of the Amazon 2. The influence
895 of geology and weathering environment on the dissolved load. *Journal of Geophysical*
896 *Research*. <https://doi.org/10.1029/JC088iC14p09671>
- 897 71. Riebe, C. S., Kirchner, J. W., & Finkel, R. C. (2004). Erosional and climatic effects on
898 long-term chemical weathering rates in granitic landscapes spanning diverse climate
899 regimes. *Earth and Planetary Science Letters*.
900 <https://doi.org/10.1016/j.epsl.2004.05.019>
- 901 72. Dixon, J. L., & von Blanckenburg, F. (2012). Soils as pacemakers and limiters of global
902 silicate weathering. *Comptes Rendus - Geoscience*.
903 <https://doi.org/10.1016/j.crte.2012.10.012>
- 904 73. Hilley, G. E., Chamberlain, C. P., Moon, S., Porder, S., & Willett, S. D. (2010).
905 Competition between erosion and reaction kinetics in controlling silicate-weathering
906 rates. *Earth and Planetary Science Letters*. <https://doi.org/10.1016/j.epsl.2010.01.008>
- 907 74. West, A. J. (2012). Thickness of the chemical weathering zone and implications for
908 erosional and climatic drivers of weathering and for carbon-cycle feedbacks. *Geology*.
909 40, 811–814. <https://doi.org/10.1130/G33041.1>
- 910 75. Caves Rügenstein, J. K., Ibarra, D. E., & von Blanckenburg, F. (2019). Neogene
911 cooling driven by land surface reactivity rather than increased weathering fluxes.
912 *Nature*. <https://doi.org/10.1038/s41586-019-1332-y>
- 913 76. Meybeck, M. (1982). Carbon, nitrogen, and phosphorus transport by world rivers.
914 *American Journal of Science*, 282(4), 401–450. <https://doi.org/10.2475/ajs.282.4.401>
- 915 77. Ludwig, W., & Probst, J. L. (1996). Predicting the oceanic input of organic carbon by
916 continental erosion. *Global Biogeochemical Cycles*.
917 <https://doi.org/10.1029/95GB02925>

- 918 78. Stallard, R. F. (1998). Terrestrial sedimentation and the carbon cycle: Coupling
919 weathering and erosion to carbon burial. *Global Biogeochemical Cycles*.
920 <https://doi.org/10.1029/98GB00741>
- 921 79. Berhe, A. A., Harte, J., Harden, J. W., & Torn, M. S. (2007). The Significance of the
922 Erosion-induced Terrestrial Carbon Sink. *BioScience*. <https://doi.org/10.1641/b570408>
- 923 80. Berner, R. A. (1982). Burial of organic carbon and pyrite sulfur in the modern ocean:
924 Its geochemical and environmental significance. *American Journal of Science*, 282(4),
925 451–473. <https://doi.org/10.2475/ajs.282.4.451>
- 926 81. Hayes, J. M., Strauss, H., & Kaufman, A. J. (1999). The abundance of ¹³C in marine
927 organic matter and isotopic fractionation in the global biogeochemical cycle of carbon
928 during the past 800 Ma. *Chemical Geology*. [https://doi.org/10.1016/S0009-2541\(99\)00083-2](https://doi.org/10.1016/S0009-2541(99)00083-2)
- 930 82. Hedges, J. I., & Keil, R. G. (1995). Sedimentary organic matter preservation: an
931 assessment and speculative synthesis. *Marine Chemistry*. [https://doi.org/10.1016/0304-4203\(95\)00008-F](https://doi.org/10.1016/0304-4203(95)00008-F)
- 933 83. Smith, R. W., Bianchi, T. S., Allison, M., Savage, C., & Galy, V. (2015). High rates of
934 organic carbon burial in fjord sediments globally. *Nature Geoscience*.
935 <https://doi.org/10.1038/NGEO2421>
- 936 84. Bianchi, T. S., Cui, X., Blair, N. E., Burdige, D. J., Eglinton, T. I., & Galy, V. (2018).
937 Centers of organic carbon burial and oxidation at the land-ocean interface. *Organic*
938 *Geochemistry*. <https://doi.org/10.1016/j.orggeochem.2017.09.008>
- 939 85. Burdige, D. J. (2007). Preservation of organic matter in marine sediments: Controls,
940 mechanisms, and an imbalance in sediment organic carbon budgets? *Chemical Reviews*.
941 <https://doi.org/10.1021/cr050347q>
- 942 86. Hilton, R. G., Galy, A., Hovius, N., Chen, M. C., Horng, M. J., & Chen, H. (2008a).
943 Tropical-cyclone-driven erosion of the terrestrial biosphere from mountains. *Nature*
944 *Geoscience*, 1(11), 759–762. <https://doi.org/10.1038/ngeo333>
- 945 87. Clark, K. E., Hilton, R. G., West, A. J., Robles Caceres, A., Gröcke, D. R., Marthews,
946 T. R., ... Malhi, Y. (2017). Erosion of organic carbon from the Andes and its effects on
947 ecosystem carbon dioxide balance. *Journal of Geophysical Research: Biogeosciences*.
948 <https://doi.org/10.1002/2016JG003615>
- 949 88. Kao, S. J., & Liu, K. K. (1996). Particulate organic carbon export from a subtropical
950 mountainous river (Lanyang Hsi) in Taiwan. *Limnology and Oceanography*.
951 <https://doi.org/10.4319/lo.1996.41.8.1749>
- 952 89. Blair, N. E., Leithold, E. L., Ford, S. T., Peeler, K. A., Holmes, J. C., & Perkey, D. W.
953 (2003). The persistence of memory: The fate of ancient sedimentary organic carbon in a
954 modern sedimentary system. *Geochimica et Cosmochimica Acta*, 67(1), 63–73.
955 [https://doi.org/10.1016/S0016-7037\(02\)01043-8](https://doi.org/10.1016/S0016-7037(02)01043-8)
- 956 **90. Galy, V., Peucker-Ehrenbrink, B., & Eglinton, T. (2015). Global carbon export**
957 **from the terrestrial biosphere controlled by erosion. *Nature*, 521(7551), 204–207.**
958 **<https://doi.org/10.1038/nature14400> — Assembled database of organic carbon**
959 **fluxes to show and quantify key relationships between erosion and organic carbon**
960 **export from the continents.**
- 961 91. Lyons, W. B., Nezat, C. A., Carey, A. E., & Hicks, D. M. (2002). Organic carbon
962 fluxes to the ocean from high-standing islands. *Geology*, 30(5), 443–446.
963 [https://doi.org/10.1130/0091-7613\(2002\)030<0443:OCFTTO>2.0.CO;2](https://doi.org/10.1130/0091-7613(2002)030<0443:OCFTTO>2.0.CO;2)

- 964 92. Hilton, R. G. (2017). Climate regulates the erosional carbon export from the terrestrial
965 biosphere. *Geomorphology*, 277, 118–132.
966 <https://doi.org/10.1016/j.geomorph.2016.03.028>
- 967 93. Hilton, R. G., Galy, A., Hovius, N., Kao, S. J., Horng, M. J., & Chen, H. (2012).
968 Climatic and geomorphic controls on the erosion of terrestrial biomass from subtropical
969 mountain forest. *Global Biogeochemical Cycles*.
970 <https://doi.org/10.1029/2012GB004314>
- 971 94. Hovius, N., Stark, C. P., Hao-Tsu, C., & Jiun-Chuan, L. (2000). Supply and removal of
972 sediment in a landslide-dominated mountain belt: Central Range, Taiwan. *Journal of*
973 *Geology*. <https://doi.org/10.1086/314387>
- 974 95. Larsen, I. J., & Montgomery, D. R. (2012). Landslide erosion coupled to tectonics and
975 river incision. *Nature Geoscience*, 5(7), 468–473. <https://doi.org/10.1038/ngeo1479>
- 976 96. Mayer, L. M. (1994). Relationships between mineral surfaces and organic carbon
977 concentrations in soils and sediments. *Chemical Geology*, 114, 347–363.
978 [https://doi.org/10.1016/0009-2541\(94\)90063-9](https://doi.org/10.1016/0009-2541(94)90063-9)
- 979 97. Hemingway, J.D., Rothman, D.H., Grant, K.E. et al. (2019). Mineral protection
980 regulates long-term global preservation of natural organic carbon. *Nature* 570, 228–
981 231. <https://doi.org/10.1038/s41586-019-1280-6>
- 982 98. Aller, R. C. (1998). Mobile deltaic and continental shelf muds as suboxic, fluidized bed
983 reactors. *Marine Geology*, 61, 143-155. [https://doi.org/10.1016/S0304-4203\(98\)00024-3](https://doi.org/10.1016/S0304-4203(98)00024-3)
984 3
- 985 99. Goldsmith, S. T., Carey, A. E., Lyons, W. B., Kao, S. J., Lee, T. Y., & Chen, J. (2008).
986 Extreme storm events, landscape denudation, and carbon sequestration: Typhoon
987 Mindulle, Choshui River, Taiwan. *Geology*. <https://doi.org/10.1130/G24624A.1>
- 988 100. Clark, K. E., West, A. J., Hilton, R. G., Asner, G. P., Quesada, C. A., Silman, M. R., ...
989 Malhi, Y. (2016). Storm-triggered landslides in the Peruvian Andes and implications
990 for topography, carbon cycles, and biodiversity. *Earth Surface Dynamics*, 4(1), 47–70.
991 <https://doi.org/10.5194/esurf-4-47-2016>
- 992 101. Hatten, J. A., Goñi, M. A., & Wheatcroft, R. A. (2012). Chemical characteristics of
993 particulate organic matter from a small, mountainous river system in the Oregon Coast
994 Range, USA. *Biogeochemistry*, 107(1–3), 43–66. <https://doi.org/10.1007/s10533-010-9529-z>
995 9529-z
- 996 102. Goñi, M. A., Hatten, J. A., Wheatcroft, R. A., & Borgeld, J. C. (2013). Particulate
997 organic matter export by two contrasting small mountainous rivers from the Pacific
998 Northwest, U.S.A. *Journal of Geophysical Research: Biogeosciences*.
999 <https://doi.org/10.1002/jgrg.20024>
- 1000 103. Wang, J., Hilton, R. G., Jin, Z., Zhang, F., Densmore, A. L., Gröcke, D. R., ... West, A.
1001 J. (2019). The isotopic composition and fluxes of particulate organic carbon exported
1002 from the eastern margin of the Tibetan Plateau. *Geochimica et Cosmochimica Acta*.
1003 <https://doi.org/10.1016/j.gca.2019.02.031>
- 1004 104. Smith, J. C., Galy, A., Hovius, N., Tye, A. M., Turowski, J. M., & Schleppe, P. (2013).
1005 Runoff-driven export of particulate organic carbon from soil in temperate forested
1006 uplands. *Earth and Planetary Science Letters*, 365, 198–208.
1007 <https://doi.org/10.1016/j.epsl.2013.01.027>
- 1008 105. West, A. J., Lin, C. W., Lin, T. C., Hilton, R. G., Liu, S. H., Chang, C. T., ... Hovius,
1009 N. (2011). Mobilization and transport of coarse woody debris to the oceans triggered by

- 1010 an extreme tropical storm. *Limnology and Oceanography*, 56(1), 77–85.
1011 <https://doi.org/10.4319/lo.2011.56.1.0077>
- 1012 106. Wohl, E., & Ogden, F. L. (2013). Organic carbon export in the form of wood during an
1013 extreme tropical storm, Upper Rio Chagres, Panama. *Earth Surface Processes and*
1014 *Landforms*. <https://doi.org/10.1002/esp.3389>
- 1015 107. Porder, S., Hilley, G. E., & Chadwick, O. A. (2007). Chemical weathering, mass loss,
1016 and dust inputs across a climate by time matrix in the Hawaiian Islands. *Earth and*
1017 *Planetary Science Letters*. <https://doi.org/10.1016/j.epsl.2007.03.047>
- 1018 108. Porder, S., Johnson, A. H., Xing, H. X., Brocard, G., Goldsmith, S., & Pett-Ridge, J.
1019 (2015). Linking geomorphology, weathering and cation availability in the Luquillo
1020 Mountains of Puerto Rico. *Geoderma*. <https://doi.org/10.1016/j.geoderma.2015.03.002>
- 1021 109. Morford, S. L., Houlton, B. Z., & Dahlgren, R. A. (2016). Geochemical and tectonic
1022 uplift controls on rock nitrogen inputs across terrestrial ecosystems. *Global*
1023 *Biogeochemical Cycles*. <https://doi.org/10.1002/2015GB005283>
- 1024 110. Hilton, R. G., Galy, A., West, A. J., Hovius, N., & Roberts, G. G. (2013). Geomorphic
1025 control on the $\delta^{15}\text{N}$ of mountain forests. *Biogeosciences*. [https://doi.org/10.5194/bg-10-](https://doi.org/10.5194/bg-10-1693-2013)
1026 [1693-2013](https://doi.org/10.5194/bg-10-1693-2013)
- 1027 111. Weintraub, S. R., Taylor, P. G., Porder, S., Cleveland, C. C., Asner, G. P., &
1028 Townsend, A. R. (2015). Topographic controls on soil nitrogen availability in a
1029 lowland tropical forest. *Ecology*. <https://doi.org/10.1890/14-0834.1>
- 1030 112. Milodowski, D. T., Mudd, S. M., & Mitchard, E. T. A. (2015). Erosion rates as a
1031 potential bottom-up control of forest structural characteristics in the Sierra Nevada
1032 Mountains. *Ecology*. <https://doi.org/10.1890/14-0649.1>
- 1033 113. Shields, G. A., & Mills, B. J. W. (2017). Tectonic controls on the long-term carbon
1034 isotope mass balance. *Proceedings of the National Academy of Sciences of the United*
1035 *States of America*. <https://doi.org/10.1073/pnas.1614506114>
- 1036 114. Copard, Y., Amiotte-Suchet, P., & Di-Giovanni, C. (2007). Storage and release of fossil
1037 organic carbon related to weathering of sedimentary rocks. *Earth and Planetary*
1038 *Science Letters*. <https://doi.org/10.1016/j.epsl.2007.03.048>
- 1039 115. Husson, J. M., & Peters, S. E. (2017). Atmospheric oxygenation driven by unsteady
1040 growth of the continental sedimentary reservoir. *Earth and Planetary Science Letters*,
1041 460, 68–75. <http://dx.doi.org/10.1016/j.epsl.2016.12.012>
- 1042 116. Keller, C. K., & Bacon, D. H. (1998). Soil respiration and georespiration distinguished
1043 by transport analyses of vadose CO_2 , $^{13}\text{CO}_2$, and $^{14}\text{CO}_2$. *Global Biogeochemical Cycles*.
1044 <https://doi.org/10.1029/98GB00742>
- 1045 117. Petsch, S. T., Berner, R. A., & Eglinton, T. I. (2000). A field study of the chemical
1046 weathering of ancient sedimentary organic matter. *Organic Geochemistry*.
1047 [https://doi.org/10.1016/S0146-6380\(00\)00014-0](https://doi.org/10.1016/S0146-6380(00)00014-0)
- 1048 118. Soulet, G., Hilton, R. G., Garnett, M. H., Dellinger, M., Croissant, T., Ogrič, M., &
1049 Klotz, S. (2018). Technical note: in situ measurement of flux and isotopic composition
1050 of CO_2 released during oxidative weathering of sedimentary rocks. *Biogeosciences*, 15,
1051 4087–4102. <https://doi.org/10.5194/bg-15-4087-2018>
- 1052 119. Galy, V., Beyssac, O., France-Lanord, C., & Eglinton, T. (2008). Recycling of graphite
1053 during Himalayan erosion: A geological stabilization of carbon in the crust. *Science*,
1054 322(5903), 943–945. <https://doi.org/10.1126/science.1161408>

- 1055 120. Bouchez, J., Beyssac, O., Galy, V., Gaillardet, J., France-Lanord, C., Maurice, L., &
1056 Moreira-Turcq, P. (2010). Oxidation of petrogenic organic carbon in the Amazon
1057 floodplain as a source of atmospheric CO₂. *Geology*. <https://doi.org/10.1130/G30608.1>
- 1058 121. Chang, S., & Berner, R. A. (1999). Coal weathering and the geochemical carbon cycle.
1059 *Geochimica et Cosmochimica Acta*. [https://doi.org/10.1016/S0016-7037\(99\)00252-5](https://doi.org/10.1016/S0016-7037(99)00252-5)
- 1060 122. White, A. F., & Buss, H. L. (2013). Natural Weathering Rates of Silicate Minerals. In
1061 *Treatise on Geochemistry: Second Edition*. [https://doi.org/10.1016/B978-0-08-095975-](https://doi.org/10.1016/B978-0-08-095975-7.00504-0)
1062 [7.00504-0](https://doi.org/10.1016/B978-0-08-095975-7.00504-0)
- 1063 123. Bolton, E. W., Berner, R. A., & Petsch, S. T. (2006). The weathering of sedimentary
1064 organic matter as a control on atmospheric O₂: II. Theoretical modeling. *American*
1065 *Journal of Science*, 306(8), 575–615. <https://doi.org/10.2475/08.2006.01>
- 1066 124. Dalai, T. K., Singh, S. K., Trivedi, J. R., & Krishnaswami, S. (2002). Dissolved
1067 rhenium in the Yamuna River System and the Ganga in the Himalaya: Role of black
1068 shale weathering on the budgets of Re, Os, and U in rivers and CO₂ in the atmosphere.
1069 *Geochimica et Cosmochimica Acta*. [https://doi.org/10.1016/S0016-7037\(01\)00747-5](https://doi.org/10.1016/S0016-7037(01)00747-5)
- 1070 125. Hilton, R. G., Galy, A., Hovius, N., Horng, M.-J., & Chen, H. (2011). Efficient
1071 transport of fossil organic carbon to the ocean by steep mountain rivers: An orogenic
1072 carbon sequestration mechanism. *Geology*, 39(1), 71–74.
1073 <https://doi.org/10.1130/G31352.1>
- 1074 126. Graz, Y., Di-Giovanni, C., Copard, Y., Mathys, N., Cras, A., & Marc, V. (2012).
1075 Annual fossil organic carbon delivery due to mechanical and chemical weathering of
1076 marly badlands areas, *Earth Surf. Process. Land.*, 37, 1263–1271,
1077 <https://doi.org/10.1002/esp.3232>
- 1078 127. Horan, K., Hilton, R. G., Selby, D., Ottley, C. J., Gröcke, D. R., Hicks, M., & Burton,
1079 K. W. (2017). Mountain glaciation drives rapid oxidation of rock-bound organic
1080 carbon. *Science Advances*. <https://doi.org/10.1126/sciadv.1701107>
- 1081 128. Hemingway, J. D., Hilton, R. G., Hovius, N., Eglinton, T. I., Haghipour, N., Wacker,
1082 L., ... Galy, V. V. (2018). Microbial oxidation of lithospheric organic carbon in rapidly
1083 eroding tropical mountain soils. *Science*. <https://doi.org/10.1126/science.aao6463>
- 1084 129. Beyssac, O., M. Simoes, J. P. Avouac, K. A. Farley, Y.-G. Chen, Y.-C. Chan, and B.
1085 Goffe' (2007), Late Cenozoic metamorphic evolution and exhumation of Taiwan,
1086 *Tectonics*, 26, TC6001, doi:10.1029/2006TC002064.
- 1087 130. Sparkes, R. B., Hovius, N., Galy, A., & Liu, J. T. (2020). Survival of graphitized
1088 petrogenic organic carbon through multiple erosional cycles. *Earth and Planetary*
1089 *Science Letters*. 531, 115992. <https://doi.org/10.1016/j.epsl.2019.115992>
- 1090 131. Petsch, S. T., Edwards, K. J., & Eglinton, T. I. (2005). Microbial transformations of
1091 organic matter in black shales and implications for global biogeochemical cycles.
1092 *Palaeogeography, Palaeoclimatology, Palaeoecology*.
1093 <https://doi.org/10.1016/j.palaeo.2004.10.019>
- 1094 132. Torres, M. A., West, A. J., Clark, K. E., Paris, G., Bouchez, J., Ponton, C., ... Adkins,
1095 J. F. (2016). The acid and alkalinity budgets of weathering in the Andes–Amazon
1096 system: Insights into the erosional control of global biogeochemical cycles. *Earth and*
1097 *Planetary Science Letters*. <https://doi.org/10.1016/j.epsl.2016.06.012>
- 1098 133. Burke, A., Present, T. M., Paris, G., Rae, E. C. M., Sandilands, B. H., Gaillardet, J., ...
1099 Adkins, J. F. (2018). Sulfur isotopes in rivers: Insights into global weathering budgets,

- 1100 pyrite oxidation, and the modern sulfur cycle. *Earth and Planetary Science Letters*.
1101 <https://doi.org/10.1016/j.epsl.2018.05.022>
- 1102 134. Das, A., Chung, C. H., & You, C. F. (2012). Disproportionately high rates of sulfide
1103 oxidation from mountainous river basins of Taiwan orogeny: Sulfur isotope evidence.
1104 *Geophysical Research Letters*. <https://doi.org/10.1029/2012GL051549>
- 1105 135. Turchyn, A. V., Tipper, E. T., Galy, A., Lo, J. K., & Bickle, M. J. (2013). Isotope
1106 evidence for secondary sulfide precipitation along the Marsyandi River, Nepal,
1107 Himalayas. *Earth and Planetary Science Letters*.
1108 <https://doi.org/10.1016/j.epsl.2013.04.033>
- 1109 136. Williamson, M. A., & Rimstidt, J. D. (1994). The kinetics and electrochemical rate-
1110 determining step of aqueous pyrite oxidation. *Geochimica et Cosmochimica Acta*.
1111 [https://doi.org/10.1016/0016-7037\(94\)90241-0](https://doi.org/10.1016/0016-7037(94)90241-0)
- 1112 137. Berner, R. A., Scott M. R., & Thomlinson, C. (1970). Carbonate alkalinity in the pore
1113 waters of anoxic marine sediments. *Limnology and Oceanography*, 15, 544–549.
- 1114 138. Winnick, M. J., Carroll, R. W. H., Williams, K. H., Maxwell, R. M., Dong, W., &
1115 Maher, K. (2017). Snowmelt controls on concentration-discharge relationships and the
1116 balance of oxidative and acid-base weathering fluxes in an alpine catchment, East
1117 River, Colorado, *Water Resour. Res.*, 53, doi:10.1002/2016WR019724.
- 1118 139. Hilton, R. G., Galy, A., & Hovius, N. (2008b). Riverine particulate organic carbon from
1119 an active mountain belt: Importance of landslides. *Global Biogeochemical Cycles*.
1120 <https://doi.org/10.1029/2006GB002905>
- 1121 140. Millot, R., Gaillardet, J., Dupré, B., & Allégre, C. J. (2003). Northern latitude chemical
1122 weathering rates: Clues from the Mackenzie River Basin, Canada. *Geochimica et*
1123 *Cosmochimica Acta*. [https://doi.org/10.1016/S0016-7037\(02\)01207-3](https://doi.org/10.1016/S0016-7037(02)01207-3)
- 1124 141. Hilton, R. G., Galy, V., Gaillardet, J., Dellinger, M., Bryant, C., O'Regan, M., ...
1125 Calmels, D. (2015). Erosion of organic carbon in the Arctic as a geological carbon
1126 dioxide sink. *Nature*. <https://doi.org/10.1038/nature14653>
- 1127 **142. Horan, K., Hilton, R. G., Dellinger, M., Tipper, E., Galy, V., Calmels, D., ...**
1128 **Burton, K. W. (2019). Carbon dioxide emissions by rock organic carbon oxidation**
1129 **and the net geochemical carbon budget of the Mackenzie River Basin. *American***
1130 ***Journal of Science*. <https://doi.org/10.2475/06.2019.02> — First study to put together**
1131 ***a comprehensive catchment-scale budget for key long-term carbon cycle fluxes, akin***
1132 ***to those in Fig. 4.***
- 1133 143. Calmels, D., Galy, A., Hovius, N., Bickle, M., West, A. J., Chen, M. C., & Chapman,
1134 H. (2011). Contribution of deep groundwater to the weathering budget in a rapidly
1135 eroding mountain belt, Taiwan. *Earth and Planetary Science Letters*.
1136 <https://doi.org/10.1016/j.epsl.2010.12.032>
- 1137 144. Lloret, E., Dessert, C., Gaillardet, J., Albéric, P., Crispi, O., Chaduteau, C., &
1138 Benedetti, M. F. (2011). Comparison of dissolved inorganic and organic carbon yields
1139 and fluxes in the watersheds of tropical volcanic islands, examples from Guadeloupe
1140 (French West Indies). *Chemical Geology*.
1141 <https://doi.org/10.1016/j.chemgeo.2010.10.016>
- 1142 145. Lloret, E., Dessert, C., Pastor, L., Lajeunesse, E., Crispi, O., Gaillardet, J., & Benedetti,
1143 M. F. (2013). Dynamic of particulate and dissolved organic carbon in small volcanic

- 1144 mountainous tropical watersheds. *Chemical Geology*, 351, 229–244.
1145 <https://doi.org/10.1016/j.chemgeo.2013.05.023>
- 1146 146. Ferguson, R. I. (1987). Accuracy and precision of methods for estimating river loads.
1147 *Earth Surface Processes and Landforms*. <https://doi.org/10.1002/esp.3290120111>
- 1148 147. Gaillardet, J., Dupré, B., & Allègre, C. J. (1999). Geochemistry of large river
1149 suspended sediments: Silicate weathering or recycling tracer? *Geochimica et*
1150 *Cosmochimica Acta*. [https://doi.org/10.1016/s0016-7037\(99\)00307-5](https://doi.org/10.1016/s0016-7037(99)00307-5)
- 1151 148. Mills, B., Daines, S. J., & Lenton, T. M. (2014). Changing tectonic controls on the
1152 long-term carbon cycle from Mesozoic to present. *Geochemistry, Geophysics,*
1153 *Geosystems*. <https://doi.org/10.1002/2014GC005530>
- 1154 149. Jenny, H. (1941). Factors of Soil Formation. *Soil Science*.
1155 <https://doi.org/10.1097/00010694-194111000-00009>
- 1156 150. Brimhall, G. H., & Dietrich, W. E. (1987). Constitutive mass balance relations between
1157 chemical composition, volume, density, porosity, and strain in metasomatic
1158 hydrochemical systems: Results on weathering and pedogenesis. *Geochimica et*
1159 *Cosmochimica Acta*. [https://doi.org/10.1016/0016-7037\(87\)90070-6](https://doi.org/10.1016/0016-7037(87)90070-6)
- 1160 151. Brantley, S. L., Buss, H., Lebedeva, M., Fletcher, R. C., & Ma, L. (2011). Investigating
1161 the complex interface where bedrock transforms to regolith. *Applied Geochemistry*.
1162 <https://doi.org/10.1016/j.apgeochem.2011.03.017>
- 1163 152. Fletcher, R. C., Buss, H. L., & Brantley, S. L. (2006). A spheroidal weathering model
1164 coupling porewater chemistry to soil thicknesses during steady-state denudation. *Earth*
1165 *and Planetary Science Letters*. <https://doi.org/10.1016/j.epsl.2006.01.055>
- 1166 153. Goodfellow, B. W., Hilley, G. E., Webb, S. M., Sklar, L. S., Moon, S., & Olson, C. A.
1167 (2016). The chemical, mechanical, and hydrological evolution of weathering granitoid.
1168 *Journal of Geophysical Research: Earth Surface*, 121(8), 1410–1435.
1169 <https://doi.org/10.1002/2016JF003822>
- 1170 154. Brantley, S. L., Lebedeva, M. I., Balashov, V. N., Singha, K., Sullivan, P. L., &
1171 Stinchcomb, G. (2017). Toward a conceptual model relating chemical reaction fronts to
1172 water flow paths in hills. *Geomorphology*.
1173 <https://doi.org/10.1016/j.geomorph.2016.09.027>
- 1174 155. Buss, H. L., Sak, P. B., Webb, S. M., & Brantley, S. L. (2008). Weathering of the Rio
1175 Blanco quartz diorite, Luquillo Mountains, Puerto Rico: Coupling oxidation,
1176 dissolution, and fracturing. *Geochimica et Cosmochimica Acta*.
1177 <https://doi.org/10.1016/j.gca.2008.06.020>
- 1178 156. Molnar, P., Anderson, R. S., & Anderson, S. P. (2007). Tectonics, fracturing of rock,
1179 and erosion. *Journal of Geophysical Research: Earth Surface*.
1180 <https://doi.org/10.1029/2005JF000433>
- 1181 157. Moon, S., Perron, J. T., Martel, S. J., Holbrook, W. S., & St. Clair, J. (2017). A model
1182 of three-dimensional topographic stresses with implications for bedrock fractures,
1183 surface processes, and landscape evolution. *Journal of Geophysical Research: Earth*
1184 *Surface*, 122(4), 823–846. <https://doi.org/10.1002/2016JF004155>
- 1185 158. Lebedeva, M. I., Fletcher, R. C., Brantley, S. L., (2010). A mathematical model for
1186 steady-state regolith production at constant erosion rate. *Earth Surface Processes and*
1187 *Landforms*, 35, 508–524. <https://doi.org/10.1002/esp.1954>
- 1188 159. Li, D. D., Jacobson, A. D., & McInerney, D. J. (2014). A reactive-transport model for
1189 examining tectonic and climatic controls on chemical weathering and atmospheric CO₂

- 1190 consumption in granitic regolith. *Chemical Geology*, 365, 30–42.
1191 <https://doi.org/10.1016/j.chemgeo.2013.11.028>
- 1192 160. Kirchner, J. W., Feng, X., & Neal, C. (2001). Catchment-scale advection and dispersion
1193 as a mechanism for fractal scaling in stream tracer concentrations. *Journal of*
1194 *Hydrology*. [https://doi.org/10.1016/S0022-1694\(01\)00487-5](https://doi.org/10.1016/S0022-1694(01)00487-5)
- 1195 161. Hilton, R. G., Meunier, P., Hovius, N., Bellingham, P. J., & Galy, A. (2011b).
1196 Landslide impact on organic carbon cycling in a temperate montane forest. *Earth*
1197 *Surface Processes and Landforms*. <https://doi.org/10.1002/esp.2191>
- 1198 162. Ramos Scharrón, C. E., Castellanos, E. J., & Restrepo, C. (2012). The transfer of
1199 modern organic carbon by landslide activity in tropical montane ecosystems. *Journal of*
1200 *Geophysical Research: Biogeosciences*. <https://doi.org/10.1029/2011JG001838>
- 1201 **163. Emberson, R., Hovius, N., Galy, A., & Marc, O. (2016). Chemical weathering in**
1202 **active mountain belts controlled by stochastic bedrock landsliding. *Nature***
1203 ***Geoscience*, 9(1), 42–45. <https://doi.org/10.1038/ngeo2600> — Revealed the**
1204 ***importance of landslides as “weathering reactors,” generating high solute fluxes that***
1205 ***can play a key role in total weathering from mountainous terrain.***
- 1206 164. Emberson, R., Hovius, N., Galy, A., & Marc, O. (2016). Oxidation of sulfides and rapid
1207 weathering in recent landslides. *Earth Surface Dynamics*. [https://doi.org/10.5194/esurf-](https://doi.org/10.5194/esurf-4-727-2016)
1208 [4-727-2016](https://doi.org/10.5194/esurf-4-727-2016)
- 1209 165. Carretier, S., Goddérès, Y., Martinez, J., Reich, M., & Martinod, P. (2018). Colluvial
1210 deposits as a possible weathering reservoir in uplifting mountains. *Earth Surface*
1211 *Dynamics*. <https://doi.org/10.5194/esurf-6-217-2018>
- 1212 166. Croissant, T., Steer, P., Lague, D., Davy, P., Jeandet, L. & Hilton, R. G. (2019) Seismic
1213 cycles, earthquakes, landslides and sediment fluxes: Linking tectonics to surface
1214 processes using a reduced-complexity model. *Geomorphology*. 339, 87-103.
1215 <https://doi.org/10.1016/j.geomorph.2019.04.017>
- 1216 167. Keefer, D. K. (1994). The importance of earthquake-induced landslides to long-term
1217 slope erosion and slope-failure hazards in seismically active regions. *Geomorphology*.
1218 [https://doi.org/10.1016/0169-555X\(94\)90021-3](https://doi.org/10.1016/0169-555X(94)90021-3)
- 1219 168. Wang, J., Jin, Z., Hilton, R. G., Zhang, F., Li, G., Densmore, A. L., ... West, A. J.
1220 (2016). Earthquake-triggered increase in biospheric carbon export from a mountain
1221 belt. *Geology*. <https://doi.org/10.1130/G37533.1>
- 1222 169. Frith, N. V., Hilton, R. G., Howarth, J. D., Gröcke, D. R., Fitzsimons, S. J., Croissant,
1223 T., ... Densmore, A. L. (2018). Carbon export from mountain forests enhanced by
1224 earthquake-triggered landslides over millennia. *Nature Geoscience*.
1225 <https://doi.org/10.1038/s41561-018-0216-3>
- 1226 170. Jin, Z., A. J. West, A., Zhang, F., An, Z., Hilton, R. G., Yu, J., ... Wang, X. (2016).
1227 Seismically enhanced solute fluxes in the Yangtze River headwaters following the A.D.
1228 2008 Wenchuan earthquake. *Geology*. <https://doi.org/10.1130/G37246.1>
- 1229 171. Kao, S. J., Dai, M. H., Wei, K. Y., Blair, N. E., & Lyons, W. B. (2008). Enhanced
1230 supply of fossil organic carbon to the Okinawa trough since the last deglaciation.
1231 *Paleoceanography*. <https://doi.org/10.1029/2007PA001440>
- 1232 172. Blair, N. E., Leithold, E. L., & Aller, R. C. (2004). From bedrock to burial: The
1233 evolution of particulate organic carbon across coupled watershed-continental margin
1234 systems. *Marine Chemistry*, 92(1-4 SPEC. ISS.), 141–156.
1235 <https://doi.org/10.1029/2002JC001467>

- 1236 173. Leithold, E. L., Blair, N. E., & Wegmann, K. W. (2016). Source-to-sink sedimentary
1237 systems and global carbon burial: A river runs through it. *Earth-Science Reviews*.
1238 <https://doi.org/10.1016/j.earscirev.2015.10.011>
- 1239 174. Aufdenkampe, A. K., Mayorga, E., Raymond, P. A., Melack, J. M., Doney, S. C., Alin,
1240 S. R., ... Yoo, K. (2011). Riverine coupling of biogeochemical cycles between land,
1241 oceans, and atmosphere. *Frontiers in Ecology and the Environment*.
1242 <https://doi.org/10.1890/100014>
- 1243 175. Torres, M. A., Limaye, A. B., Ganti, V., Lamb, M. P., West, A. J., and Fischer, W. W.
1244 (2017) Model predictions of long-lived storage of organic carbon in river deposits,
1245 *Earth Surface Dynamics*, 5, 711–730, <https://doi.org/10.5194/esurf-5-711-2017>, 2017.
- 1246 176. Galy, V., Eglinton, T., France-Lanord, C., & Sylva, S. (2011). The provenance of
1247 vegetation and environmental signatures encoded in vascular plant biomarkers carried
1248 by the Ganges-Brahmaputra rivers. *Earth and Planetary Science Letters*.
1249 <https://doi.org/10.1016/j.epsl.2011.02.003>
- 1250 177. Feakins, S. J., Wu, M. S., Ponton, C., Galy, V., & West, A. J. (2018). Dual isotope
1251 evidence for sedimentary integration of plant wax biomarkers across an Andes-Amazon
1252 elevation transect. *Geochimica et Cosmochimica Acta*.
1253 <https://doi.org/10.1016/j.gca.2018.09.007>
- 1254 178. Ponton, C., West, A. J., Feakins, S. J., & Galy, V. (2014). Leaf wax biomarkers in
1255 transit record river catchment composition. *Geophysical Research Letters*.
1256 <https://doi.org/10.1002/2014GL061328>
- 1257 179. Hemingway, J. D., Schefuß, E., Spencer, R. G. M., Bienvenu, J. D., Eglinton, T. I.,
1258 McIntyre, C., Galy, V. V. (2017). Hydrologic controls on seasonal and inter-annual
1259 variability of Congo River particulate organic matter source and reservoir age.
1260 *Chemical Geology*, 466, 454-465. <https://doi.org/10.1016/j.chemgeo.2017.06.034>
- 1261 180. Scheingross, J. S., Hovius, N., Dellinger, M., Hilton, R. G., Repasch, M., Sachse, D.,
1262 ... Turowski, J. M. (2019). Preservation of organic carbon during active fluvial
1263 transport and particle abrasion. *Geology*. <https://doi.org/10.1130/G46442.1>
- 1264 181. Johnson, J. E., Gerpheide, A., Lamb, M. P., & Fischer, W. W. (2014). O₂ constraints
1265 from Paleoproterozoic detrital pyrite and uraninite. *Bulletin of the Geological Society of*
1266 *America*. <https://doi.org/10.1130/B30949.1>
- 1267 182. Dellinger, M., Gaillardet, J., Bouchez, J., Calmels, D., Louvat, P., Dosseto, A.,
1268 ... Maurice, L. (2015). Riverine Li isotope fractionation in the Amazon River basin
1269 controlled by the weathering regimes. *Geochimica et Cosmochimica Acta*.
1270 <https://doi.org/10.1016/j.gca.2015.04.042>
- 1271 183. Lupker, M., France-Lanord, C., Galy, V., Lavé, J., Gaillardet, J., Gajurel, A. P., ...
1272 Sinha, R. (2012). Predominant floodplain over mountain weathering of Himalayan
1273 sediments (Ganga basin). *Geochimica et Cosmochimica Acta*.
1274 <https://doi.org/10.1016/j.gca.2012.02.001>
- 1275 184. Bickle, M. J., Chapman, H. J., Tipper, E., Galy, A., De La Rocha, C. L., & Ahmad, T.
1276 (2018). Chemical weathering outputs from the flood plain of the Ganga. *Geochimica et*
1277 *Cosmochimica Acta*. <https://doi.org/10.1016/j.gca.2018.01.003>
- 1278 185. Bouchez, J., Gaillardet, J. Ô., Lupker, M., Louvat, P., France-Lanord, C., Maurice, L.,
1279 ... Moquet, J. S. (2012). Floodplains of large rivers: Weathering reactors or simple
1280 silos? *Chemical Geology*. <https://doi.org/10.1016/j.chemgeo.2012.09.032>

- 1281 186. Moquet, J. S., Guyot, J. L., Crave, A., Viers, J., Filizola, N., Martinez, J. M., ...
1282 Pombosa, R. (2016). Amazon River dissolved load: temporal dynamics and annual
1283 budget from the Andes to the ocean. *Environmental Science and Pollution Research*.
1284 <https://doi.org/10.1007/s11356-015-5503-6>
- 1285 187. Lupker, M., France-Lanord, C., Lavé, J., Bouchez, J., Galy, V., Métivier, F., ...
1286 Mugnier, J. L. (2011). A Rouse-based method to integrate the chemical composition of
1287 river sediments: Application to the Ganga basin. *Journal of Geophysical Research:*
1288 *Earth Surface*. <https://doi.org/10.1029/2010JF001947>
- 1289 188. Lupker, M., France-Lanord, C., Galy, V., Lavé, J. Ô., & Kudrass, H. (2013). Increasing
1290 chemical weathering in the Himalayan system since the Last Glacial Maximum. *Earth*
1291 *and Planetary Science Letters*. <https://doi.org/10.1016/j.epsl.2013.01.038>
- 1292 189. Clift, P. D., Hodges, K. V., Heslop, D., Hannigan, R., Van Long, H., & Calves, G.
1293 (2008). Correlation of Himalayan exhumation rates and Asian monsoon intensity.
1294 *Nature Geoscience*, 1(12), 875–880. <https://doi.org/10.1038/ngeo351>
- 1295 190. Wan, S., Clift, P. D., Li, A., Yu, Z., Li, T., & Hu, D. (2012). Tectonic and climatic
1296 controls on long-term silicate weathering in Asia since 5 Ma. *Geophysical Research*
1297 *Letters*, 39(15). <https://doi.org/10.1029/2012GL052377>
- 1298 191. Frings, P. J. (2019). Palaeoweathering: How Do Weathering Rates Vary with Climate?
1299 *Elements*. <https://doi.org/10.2138/gselements.15.4.259>
- 1300 192. Schachtman, N. S., Roering, J. J., Marshall, J. A., Gavin, D. G., & Granger, D. E.
1301 (2019). The interplay between physical and chemical erosion over glacial-interglacial
1302 cycles. *Geology*. <https://doi.org/10.1130/G45940.1>
- 1303 193. Peucker-Ehrenbrink, B., & Ravizza, G. (2000). The marine osmium isotope record.
1304 *Terra Nova*. <https://doi.org/10.1046/j.1365-3121.2000.00295.x>
- 1305 194. Li, G., & Elderfield, H. (2013). Evolution of carbon cycle over the past 100 million
1306 years. *Geochimica et Cosmochimica Acta*. <https://doi.org/10.1016/j.gca.2012.10.014>
- 1307 195. Misra, S., & Froelich, P. N. (2012). Lithium isotope history of cenozoic seawater:
1308 Changes in silicate weathering and reverse weathering. *Science*.
1309 <https://doi.org/10.1126/science.1214697>
- 1310 196. Willenbring, J. K., & Von Blanckenburg, F. (2010). Long-term stability of global
1311 erosion rates and weathering during late-Cenozoic cooling. *Nature*.
1312 <https://doi.org/10.1038/nature09044>
- 1313 197. Foster, G. L., Royer, D. L., & Lunt, D. J. (2017). Future climate forcing potentially
1314 without precedent in the last 420 million years. *Nature Communications*.
1315 <https://doi.org/10.1038/ncomms14845>
- 1316 198. Shackleton, N. J. (1987). The carbon isotope record of the Cenozoic: History of organic
1317 carbon burial and of oxygen in the ocean and atmosphere. *Geological Society Special*
1318 *Publication*. <https://doi.org/10.1144/GSL.SP.1987.026.01.27>
- 1319 199. Derry, L. A. (2013). Organic Carbon Cycling and the Lithosphere. In *Treatise on*
1320 *Geochemistry: Second Edition*. <https://doi.org/10.1016/B978-0-08-095975-7.01014-7>
- 1321 200. Mason, E., Edmonds, M., & Turchyn, A. V. (2017). Remobilization of crustal carbon
1322 may dominate volcanic arc emissions. *Science*, 357(6348), 290–294.
1323 <https://doi.org/10.1126/science.aan5049>
- 1324 201. Dessert, C., Dupré, B., Gaillardet, J., François, L. M., & Allègre, C. J. (2003). Basalt
1325 weathering laws and the impact of basalt weathering on the global carbon cycle.
1326 *Chemical Geology*. <https://doi.org/10.1016/j.chemgeo.2002.10.001>

- 1327 202. Rad, S., Rive, K., Vittecoq, B., Cerdan, O., & Allegre, C. J. (2013). Chemical
1328 weathering and erosion rates in the Lesser Antilles: An overview in Guadeloupe,
1329 Martinique and Dominica. *Journal of South American Earth Sciences*, 45, 331-344.
1330 <Go to ISI>://WOS:000320216900022
- 1331 203. Borker, J., Hartmann, J., Romero-Mujalli, G., & Li, G. J. (2019). Aging of basalt
1332 volcanic systems and decreasing CO₂ consumption by weathering. *Earth Surface*
1333 *Dynamics*, 7(1), 191-197. <Go to ISI>://WOS:000457841200001
- 1334 204. Macdonald, F. A., Swanson-Hysell, N. L., Park, Y., Lisiecki, L., & Jagoutz, O. (2019).
1335 Arc-continent collision in tropics set Earth's climate state. *Science*, 364, 181-184.
1336 <https://doi.org/10.1126/science.aav5300>
- 1337 205. Wilson, J. T. (1968). Static or mobile earth: The current scientific revolution.
1338 *Tectonophysics*. [https://doi.org/10.1016/0040-1951\(69\)90033-x](https://doi.org/10.1016/0040-1951(69)90033-x)
- 1339 206. Blattmann, T. M., Wang, S. L., Lupker, M., Marki, L., Haghypour, N., Wacker, L.,
1340 Chung, L. H., Bernasconi, S. M., Plotze, M., & Eglinton, T. I. (2019). Sulphuric acid-
1341 mediated weathering on Taiwan buffers geological atmospheric carbon sinks. *Scientific*
1342 *Reports*, 9, 2945, <https://doi.org/10.1038/s41598-019-39272-5>
- 1343 207. White, A. F., & Blum, A. E. (1995). Effects of climate on chemical weathering in
1344 watersheds. *Geochimica et Cosmochimica Acta*. [https://doi.org/10.1016/0016-](https://doi.org/10.1016/0016-7037(95)00078-E)
1345 [7037\(95\)00078-E](https://doi.org/10.1016/0016-7037(95)00078-E)
- 1346 208. Syvitski, J. P. M., Vörösmarty, C. J., Kettner, A. J., & Green, P. (2005). Impact of
1347 humans on the flux of terrestrial sediment to the global coastal ocean. *Science*.
1348 <https://doi.org/10.1126/science.1109454>
- 1349 209. Kao, S. J., & Milliman, J. D. (2008). Water and sediment discharge from small
1350 mountainous rivers, Taiwan: The roles of lithology, episodic events, and human
1351 activities. *Journal of Geology*. <https://doi.org/10.1086/590921>
- 1352 210. Gomez, B., Carter, L., & Trustrum, N. A. (2007). A 2400 yr record of natural events
1353 and anthropogenic impacts in intercorrelated terrestrial and marine sediment cores:
1354 Waipaoa sedimentary system, New Zealand. *Bulletin of the Geological Society of*
1355 *America*. <https://doi.org/10.1130/B25996.1>
- 1356 211. Ross, M. R. V., Nippgen, F., Hassett, B. A., McGlynn, B. L., & Bernhardt, E. S.
1357 (2018). Pyrite Oxidation Drives Exceptionally High Weathering Rates and Geologic
1358 CO₂ Release in Mountaintop-Mined Landscapes. *Global Biogeochemical Cycles*.
1359 <https://doi.org/10.1029/2017GB005798>
- 1360 212. Regnier, P., Friedlingstein, P., Ciais, P., Mackenzie, F. T., Gruber, N., Janssens, I. A.,
1361 ... Thullner, M. (2013). Anthropogenic perturbation of the carbon fluxes from land to
1362 ocean. *Nature Geoscience*. <https://doi.org/10.1038/ngeo1830>
- 1363 **213. Zolkos, S., Tank, S. E., & Kokelj, S. V. (2018). Mineral Weathering and the**
1364 **Permafrost Carbon-Climate Feedback. *Geophysical Research Letters*.**
1365 **<https://doi.org/10.1029/2018GL078748> — Documented large increases in sulfide**
1366 **oxidation and associated CO₂ release associated with thawing permafrost slumps in**
1367 **the Canadian Arctic, pointing to potential weathering-driven positive feedbacks**
1368 **associated with warming.**
- 1369 214. Lyons, S.L., Baczynski, A.A., Babila, T.L. et al. (2019). Palaeocene–Eocene Thermal
1370 Maximum prolonged by fossil carbon oxidation. *Nature Geoscience*, 12, 54–60.
1371 <https://doi.org/10.1038/s41561-018-0277-3>

- 1372 215. Quéré, C., Andrew, R., Friedlingstein, P., Sitch, S., Hauck, J., Pongratz, J., ... Zheng,
1373 B. (2018). Global Carbon Budget 2018. *Earth System Science Data*.
1374 <https://doi.org/10.5194/essd-10-2141-2018>
- 1375 216. Archer, D., Eby, M., Brovkin, V., Ridgwell, A., Cao, L., Mikolajewicz, U., ... Tokos,
1376 K. (2009). Atmospheric Lifetime of Fossil Fuel Carbon Dioxide. *Annual Review of*
1377 *Earth and Planetary Sciences*. <https://doi.org/10.1146/annurev.earth.031208.100206>
- 1378 217. Norton, K. P., & von Blanckenburg, F. (2010). Silicate weathering of soil-mantled
1379 slopes in an active Alpine landscape. *Geochimica et Cosmochimica Acta*.
1380 <https://doi.org/10.1016/j.gca.2010.06.019>
- 1381 218. Dixon, J. L., Hartshorn, A. S., Heimsath, A. M., DiBiase, R. A., & Whipple, K. X.
1382 (2012). Chemical weathering response to tectonic forcing: A soils perspective from the
1383 San Gabriel Mountains, California. *Earth and Planetary Science Letters*.
1384 <https://doi.org/10.1016/j.epsl.2012.01.010>

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1386 **Key points:**

- 1387 - Erosion resulting from mountain building increases transfer of carbon between the
1388 atmosphere and storage in rocks
- 1389 - The traditional view has focused on CO₂ drawdown by silicate weathering, and its
1390 links to climate and erosion
- 1391 - An emerging view also considers CO₂ drawdown by organic carbon burial, and CO₂
1392 emissions from oxidative weathering of both rock organic carbon and sulfide minerals
- 1393 - CO₂ sources and sinks increase with erosion, and the net balance has now been
1394 quantified in a handful of locations
- 1395 - Climate (temperature, hydrology) regulates inorganic and organic CO₂ sinks, with
1396 complex interdependency on erosion
- 1397 - Lithology is important: a mountain range composed of sedimentary rocks may be a
1398 weak CO₂ sink (or CO₂ source), but volcanic rocks favor CO₂ drawdown

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1407

1408 **Author contributions:**

1409 RGH and AJW formulated the review and identified the themes to be covered. RGH
1410 compiled the datasets and drafted the figures. RGH and AJW contributed equally to the
1411 discussion and writing of the manuscript.

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1414 **Competing interests:**

1415 The authors declare that they have no competing interests.

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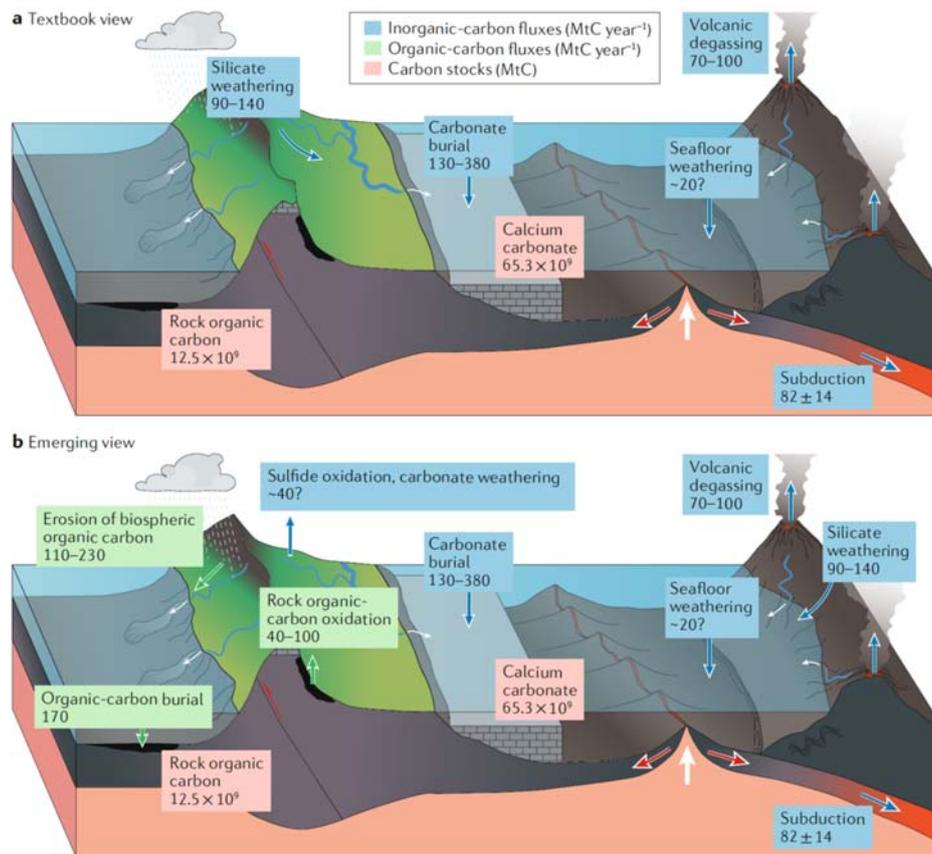
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1423 **Supplementary information:**

1424 Supplementary information is available for this paper at the end of the document.

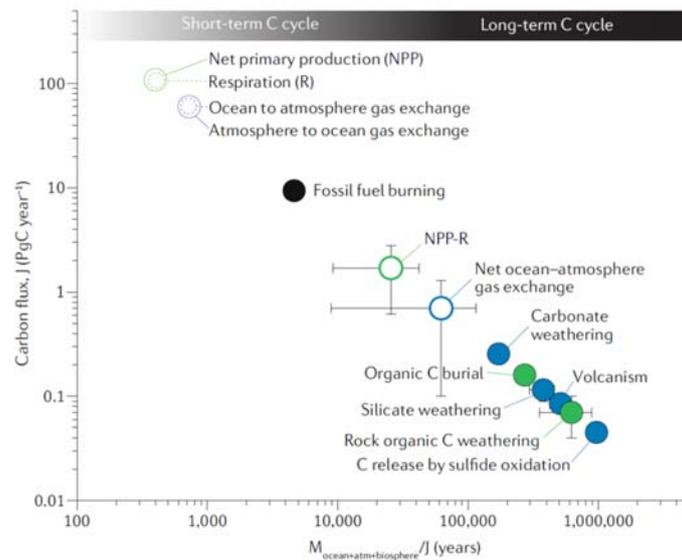
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1427 **Figure 1: The geological carbon cycle and transfers of carbon between the atmosphere**
1428 **and rocks. A.** The textbook view³⁵ that juxtaposes CO₂ emissions from volcanism^{44,45} against
1429 silicate mineral weathering and carbonate burial (inorganic carbon transfers shown in
1430 blue)^{18,51}. **B.** The emerging view that we highlight in this Review, which also considers the
1431 organic carbon pathways shown in green for: the erosion of the terrestrial biosphere and
1432 transfer by rivers⁹⁰, organic carbon burial in the ocean^{82,83,85} and the oxidative weathering of
1433 rock organic carbon⁴⁰. Additional CO₂ release can occur as a result of sulfide oxidation^{132,133}.
1434 Carbon stocks in rocks as carbonate minerals and rock organic carbon are provided in
1435 Megatonnes of carbon (MtC)⁴⁹. Carbon fluxes that operate on short timescales (< 100s of
1436 kyrs, such as photosynthesis, respiration, and carbonate weathering; Fig. 2) are not shown.

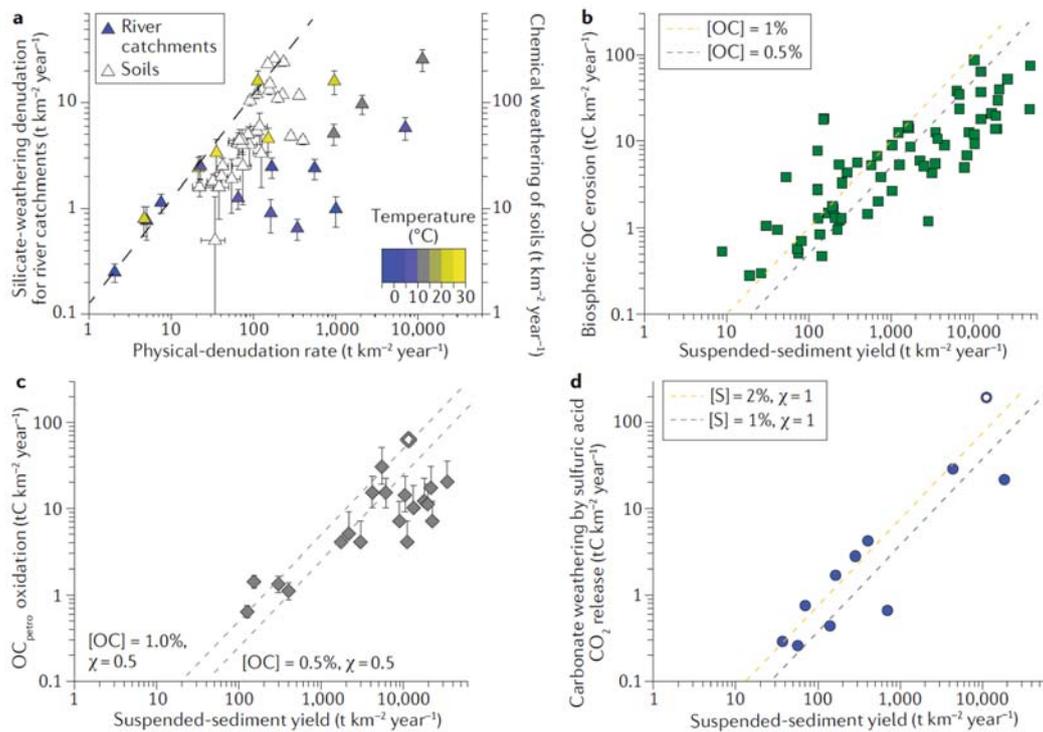
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1439 **Figure 2: Comparison of major fluxes and timescales of relevance in the global carbon**
 1440 **cycle.** Estimated pre-industrial fluxes of carbon (J) (refs.^{48, 49}) are shown along with the
 1441 timescale required for each flux to replace the entire carbon mass of the oceans, atmosphere
 1442 and biosphere ($M_{\text{ocean+atm+biosphere}} = 43,540 \pm 550 \text{ PgC}$). Green symbols denote the organic
 1443 carbon cycle, and blue the inorganic carbon cycle. Open circles show processes that dominate
 1444 the ‘short-term’ carbon cycle (<100,000 years), which include net primary production on land
 1445 and in the ocean, respiration, and ocean-atmosphere gas exchanges. The net difference
 1446 between the short-term fluxes (“NPP – R”, and “Net ocean-atmosphere gas exchange”) can
 1447 control the carbon cycle over decades to thousands of years. The processes that dominate the
 1448 long-term carbon cycle, over 10^5 to 10^6 year timescales, are shown as filled circles. The rapid
 1449 acceleration of rock organic carbon oxidation by fossil fuel burning is highlighted as a black
 1450 symbol.

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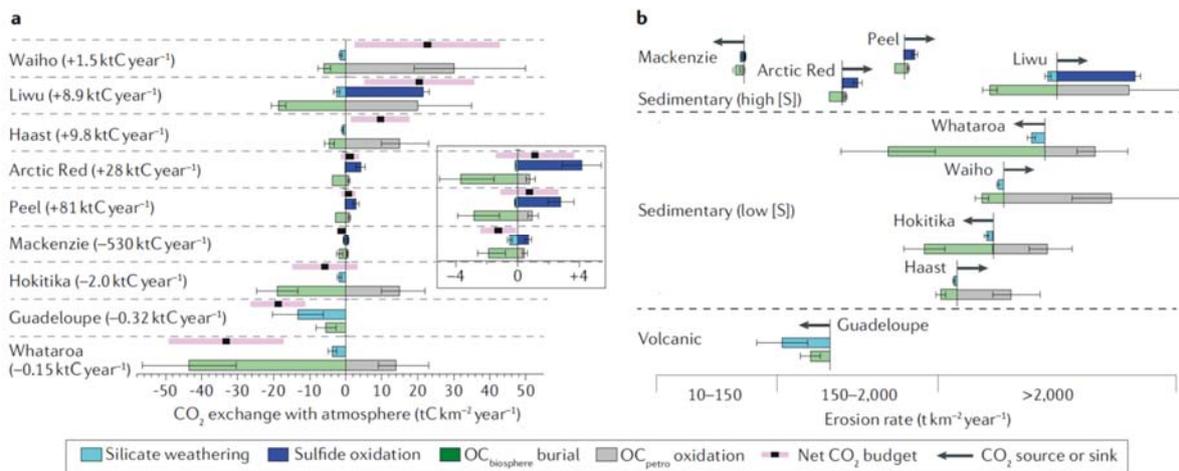
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1453 **Figure 3: A river catchment view of physical erosion rate versus carbon transfer. A)**

1454 Relationship between physical denudation rate and silicate weathering fluxes. Coloured
1455 symbols refer to the cation flux derived from silicate weathering for selected river catchments
1456 draining felsic rocks³², with colour corresponding to mean annual temperature. Open symbols
1457 represent measurements of chemical weathering fluxes determined from granitic soils^{72,217,218}.
1458 The river catchment silicate weathering rates are ~10% of the soil-derived total chemical
1459 weathering rates, consistent with the observation that there are ~10% silicate cations in felsic
1460 rocks³². The black line has a gradient of 1, representing a linear trend and showing a supply-
1461 limited control on silicate weathering fluxes. **B)** Erosion of biospheric organic carbon (OC)
1462 versus the suspended sediment yield in rivers globally. The erosional flux of biospheric OC is
1463 determined using stable isotopes and radiocarbon (to separate biospheric and petrogenic
1464 OC)^{87,90,92,103}. Dotted lines indicate the trends predicted if the eroded sediments contain a
1465 constant concentration of OC. **C)** Oxidation of rock-derived organic carbon (OC_{petro})
1466 calculated using rhenium as a proxy^{41,124,127,142} (closed symbols) and direct measurements¹¹⁸
1467 (open symbol). Dotted lines represent expected weathering rates of various rock organic
1468 carbon contents and a weathering intensity of 50% ($\chi=0.5$). Compiled data are provided in the
1469 Supplementary Information. **D)** CO₂ release associated with sulfide oxidation, assuming a 1:1
1470 molar ratio release of sulfur and carbon resulting from sulfuric acid weathering of carbonates.
1471 Closed symbols represent estimates from river chemistry^{19,42,43,134} and open symbols from
1472 direct measurements¹¹⁸. Dotted lines indicate weathering rates assuming a constant
1473 weathering intensity (complete oxidation, $\chi=1$) for given sulfur content in bedrocks.

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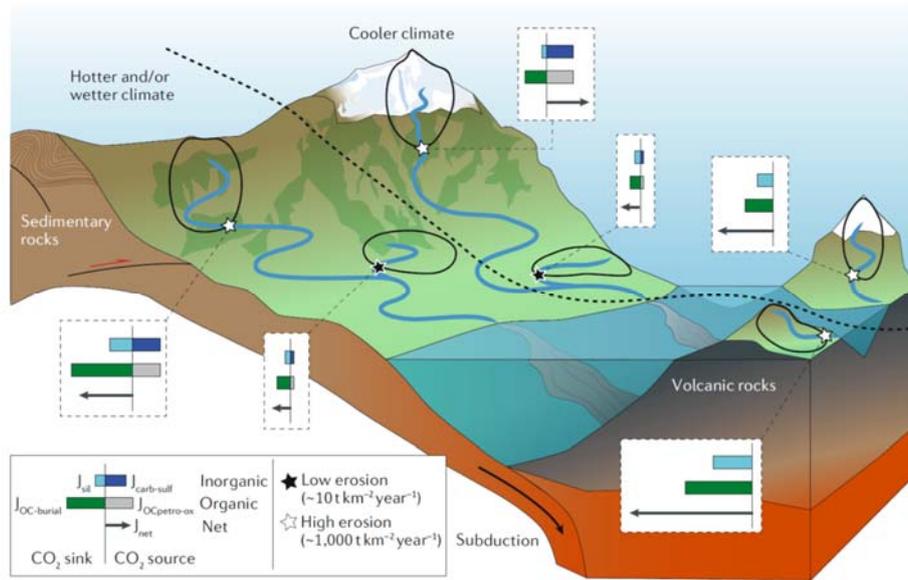
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Figure 4: Net “rock-atmosphere” CO₂ exchange in river catchments. **A.** The potential CO₂ sinks by silicate weathering (light blue) and biospheric organic carbon (OC_{biosphere}) burial (green) are shown as negative fluxes. CO₂ sources via oxidative weathering of rock-derived organic carbon (OC_{petro}; grey) and sulfides (dark blue) are shown as positive fluxes. Uncertainties on all fluxes are provided as whiskers. The net geochemical carbon flux in each catchment is given as a black tick (uncertainty range provided by pink bars). Numbers in parentheses beside catchment names represent the net transfer of CO₂ in ktC yr⁻¹. Inset shows magnification of catchments with lower yields. **B.** CO₂ sources and sinks shown by the same coloured bars as in part A, but the catchments are arranged in terms of lithology and physical erosion rate. The black arrow indicates whether the net CO₂ exchange is a source or sink (refer to part A for the absolute values). Compiled data^{41-43,69,93,139-145} are provided in the Supplementary Information.



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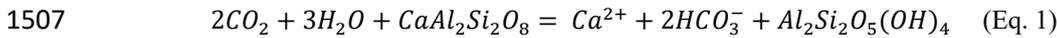
1490 **Figure 5: A new view of mountains, erosion and the carbon cycle.** Schematic of the CO₂
 1491 sources and sinks by weathering in different environments, including fluxes in the inorganic
 1492 carbon cycle (J_{sil} = silicate weathering, light blue; $J_{\text{carb-sulf}}$ = sulfide oxidation, dark blue) and
 1493 organic carbon cycle (J_{OCburial} = OC burial, dark green; $J_{\text{OCpetro-ox}}$ = rock organic carbon
 1494 oxidation, grey), along with the net effect of “rock-atmosphere” fluxes on CO₂ (black arrow).
 1495 The cartoon considers catchments with high erosion rates, where reactions may be
 1496 “kinetically limited”, and low erosion rates, where reactions are “supply limited”, shown by
 1497 the white and black stars, respectively. The influence of lithology is shown by sedimentary
 1498 rock-dominated catchments on the left side, and catchments underlain by volcanic rocks on
 1499 the right. The dotted line separates: a) hotter or wetter climates on the near side; and b) cooler
 1500 climates on the on the far side. Stylized fluxes represent qualitative estimates from these
 1501 different environments based on data collected to date (Fig. 4).

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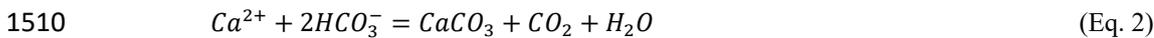
1503 **Box 1: CO₂ sources and sinks by weathering and erosion**

1504 *1. Silicate weathering as a CO₂ sink*

1505 Dissolution of atmospheric CO₂ in rainwater (H₂O) leads to the formation of carbonic acid, which causes the
1506 breakdown of silicate minerals via:

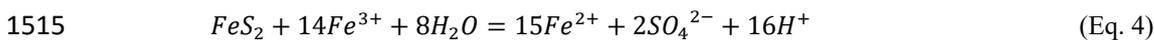
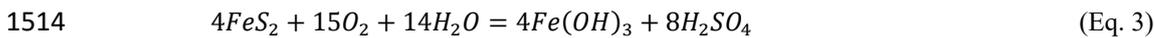


1508 Silicate weathering reactions result in long-term CO₂ drawdown when the reaction products are used to form
1509 calcium carbonate (CaCO₃), following:

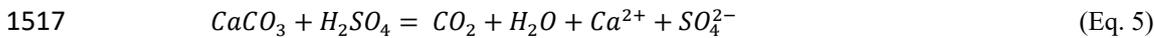


1511 *2. Sulfide oxidation as a CO₂ source*

1512 Sulfide minerals (such as pyrite, FeS₂) can be oxidised by gaseous or dissolved O₂ (Eq. 3), or by reduction of
1513 another oxidised species (such as ferric iron, Fe³⁺; Eq. 4):



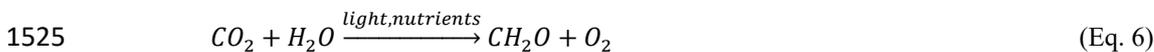
1516 Sulfuric acid (produced in both cases) can weather carbonate minerals (Eq. 1), following:



1518 thus acting as a source of CO₂. Alternatively, the production of sulfuric acid might alter the alkalinity balance of
1519 natural waters, also resulting in CO₂ release. Either way, the CO₂ released in association with sulfide oxidation
1520 is balanced by alkalinity production during sulfate reduction and sulfide precipitation in marine sediments.
1521 However, the long residence time of SO₄²⁻ in the oceans (~ 10 Myrs at present) means that sulfide oxidation
1522 might have a persistent effect on atmospheric CO₂ concentration^{42,43}.

1523 *3. Biospheric POC erosion as a CO₂ sink*

1524 Photosynthesis forms organic matter, removing CO₂ from the atmosphere and producing O₂:



1526 Erosion can export particulate organic carbon (POC) from forests and deliver it to rivers⁷⁶. If this POC is buried
1527 in sediments⁸⁰, and the eroded terrestrial OC is replaced by new photosynthetic production⁷⁹, the net effect is
1528 long-term CO₂ drawdown through burial of OC in rocks.

1529 *4. Oxidation of OC_{petro} as a CO₂ source*

1530 When sedimentary rocks are exhumed, OC_{petro} (also described as ‘fossil’ OC⁸⁸) can break down chemically as:



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1532 There is substantial evidence that respiration of OC_{petro}, which might be mediated by microbiology, can occur in
1533 the weathering zone^{116,118,128,131}.

1534

1535 **Glossary**

1536 **Biospheric organic carbon** – Carbon derived from living plants and degraded organic matter
1537 in soils, up to a few thousands of years in age.

1538 **Critical zone** – The region from the top of the vegetation canopy to the base of the ground
1539 water; where rocks, water, atmosphere and life interact.

1540 **Weathering flux** – The rate of the mass transfer of weathering products ($t\ km^{-2}\ yr^{-1}$).
1541 Equivalent to the product of total denudation rate and chemical weathering intensity.

1542 **Weathering intensity** – The ratio between chemical denudation and total denudation (
1543 represented by a fraction or percent).

1544 **Denudation** – The total loss of mass from a landscape, driven by erosion (physical
1545 denudation) and/or by chemical weathering (chemical denudation; $t\ km^{-2}\ yr^{-1}$).

1546 **Draw down of carbon dioxide** – Transfer of C from the CO₂ molecule in the atmosphere to
1547 bicarbonate, carbonate, or organic matter.

1548 **Erosion** – The movement of mass across Earth's surface, usually by fluids (granular, liquid,
1549 or gas; $t\ km^{-2}\ yr^{-1}$).

1550 **Landslides** – An erosion process that acts to move material in a rapid motion and results in
1551 transfer of mass downslope.

1552 **Mountain building** – The formation of a mountain range due to tectonic plate convergence,
1553 folding and faulting, or through dynamic forces that act on Earth's crust.

1554 **Petrogenic OC** – Organic carbon that is rock-derived, typically defined on the basis of being
1555 depleted in radiocarbon (therefore older than $\sim >60,000$ years).

1556 **Reactivity** – The tendency of a substance (atom, molecule) to undergo a reaction; is
1557 considered in terms of the individual phase (silicate mineral, organic molecule) and
1558 associated acid-base or reduction/oxidation reactions in chemical weathering.

1559 **Shales** – A type of sedimentary rock that is typically fine grained and mostly made up of silt
1560 and clay sized clasts, and can contain up to a few weight percent of carbonate, OC_{petro} and
1561 sulfide minerals.

1562 **“Supply limited” weathering** - when chemical weathering reactions are limited by the
1563 supply of minerals to react.

1564 **Chemical weathering** – The chemical processes that disintegrate (break up, loosen) rock,
1565 altering its original characteristics and producing weathering products.

1566 **Weathering front** – A marked gradient in the chemical composition of a weathering profile
1567 where a parameter changes from the original un-weathered rock, to the solid weathering
1568 products.

1569 **Weathering limited:** when chemical weathering fluxes are limited by factors that control the
1570 rate of reaction, such as temperature and fluid flow

1571 **Weathering profile** – A one dimensional view of the chemical and/or physical changes to
1572 rocks as they are exposed to life, water and the atmosphere.

1573 **Weathering thermostat** – The response of weathering fluxes to changes in climate that act
1574 to stabilise atmospheric CO₂ and Earth’s surface temperature; increases in temperature and/or
1575 CO₂ concentrations cause a response that acts to draw down CO₂.

1576

1577 **Summary:**

1578 By increasing erosion, mountain building can steer the evolution of atmospheric CO₂ and
1579 global climate. This Review expands from the canonical focus on erosion and silicate
1580 weathering to consider the net carbon budget of erosion, including both CO₂ sinks (silicate
1581 weathering, organic carbon burial) and CO₂ sources (oxidative weathering) together.