

1 **Forearc carbon sequestration reduces long-term volatile recycling into the mantle**

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55 **Summary:**

56 Carbon and other volatiles are transported from Earth's surface into the
57 mantle at subduction margins. The efficiency of this transfer has profound
58 implications for the nature and scale of geochemical heterogeneities in Earth's deep
59 (mantle) and shallow (crustal) reservoirs, as well as Earth's oxidation state. However,
60 the proportion of volatiles released in the forearc and backarc are not well-
61 constrained compared to fluxes from the volcanic-front. Here, we use helium and
62 carbon isotope data from deeply-sourced springs along two cross-arc transects to
63 show that ~91% of carbon released from the slab/mantle beneath the Costa Rica
64 forearc is sequestered within the crust by calcite deposition, and an additional ~3% is
65 incorporated into biomass through microbial chemolithoautotrophy. We estimate
66 that $\sim 1.2 \times 10^8$ to 1.3×10^{10} mol CO₂/yr are released from the slab beneath the
67 forearc, resulting in up to ~19% less carbon being transferred to Earth's deep mantle
68 than previously estimated.

69

70 **Main Text:**

71 Terrestrial carbon traverses various reservoirs on Earth from the surface to the
72 mantle. It is subjected to a number of geological, geochemical, and biological cycles, each
73 of which operates on vastly different temporal and spatial scales. Perhaps the most
74 important physical process linking the deep and shallow carbon cycles is subduction,
75 which transports both oxidized and reduced forms of crustal carbon into the mantle.
76 During subduction, volatile-rich fluids are released from the downgoing slab (upper
77 mantle and crust). These fluids are thought to migrate through the overlying mantle-
78 wedge and crust, and are ultimately released across the forearc, volcanic arc-front and
79 backarc (i.e., behind the volcanic front). However, little is known about what effect
80 volatile fluxes through the forearc may have on the total volatile budget and estimates of
81 recycling efficiency¹. Much of the forearc subsurface is at low temperatures (<100°C) that
82 are conducive to microbial life as well as shallow water-rock interactions. Therefore,

83 microorganisms and/or abiotic chemical processes may alter the speciation, isotopic
84 composition, and presumed carbon budgets. Regional-scale effects of these shallow
85 processes on volatile fluxes in the forearc have not previously been quantified.

86

87 Carbon budgets for convergent margins typically assume negligible carbon
88 emissions from the area between the trench (i.e., where the downgoing slab subducts
89 beneath the overriding plate) and the degassing volcanic arc-front¹⁻³. It is not clear
90 whether the lack of obvious high emission sources reflects a lack of deep CO₂ input from
91 the slab-mantle mixture, or if secondary processes in the upper plate⁴⁻⁵ mask diffuse but
92 significant CO₂ release.

93

94 Carbon outputs at the Central American Volcanic Arc, which have been the focus
95 of several studies, are estimated to represent only 12% of the total subducted carbon
96 input along the middle American trench^{2,6-7}, whereas global arcs typically have carbon
97 outputs that approach their inputs, suggesting limited volatile delivery to the mantle¹.
98 However, recent work⁸ proposed a far higher volcanic carbon flux for Central America,
99 suggesting that this region may be more similar to other arc segments¹ than previously
100 thought^{2,6}. Our study focuses on the Nicoya Peninsula area of Costa Rica in part because it
101 is one of the few places on Earth with easy sampling access to an on-land forearc region
102 and thus presents an opportunity to study the fate of carbon across an entire arc. Few
103 carbon flux estimates have been made from the forearc region of any arc, despite the fact
104 that there is plentiful ancillary evidence for fluid venting in forearc regions (*e.g.*,
105 serpentinite diapirism in the Marianas⁹ and mud volcanism at various accretionary prisms
106 worldwide¹⁰), as well as ample geophysical evidence that shows fluid upwelling in the
107 forearc region from the downgoing slab¹¹. In Costa Rica, the only available forearc data
108 are from three submarine venting sites³, where the authors estimated that the carbon
109 output from the outer forearc represents less than 1% of the carbon input at the trench.
110 Taking into account the loss at the volcanic-front, they proposed two explanations: either
111 subducted carbon was largely transported into the deep mantle or significant quantities

112 of carbon were lost to the forearc or backarc regions. Independently, thermodynamic
113 models of subduction predict significant CO₂ loss (i.e., decarbonation) from the top of the
114 subducting slab beneath global forearc regions¹², as the slab penetrates to greater
115 temperature and pressure conditions.

116

117 The extensive occurrence of calcite veining throughout the Costa Rica forearc¹³⁻¹⁴
118 suggests that much of the CO₂ released from the slab beneath the forearc could be
119 sequestered into the crust as calcite, in which case direct measurements of diffuse
120 degassing in this region are not representative of deep mantle processes. Additional
121 mechanisms for masking CO₂ gas release in the upper plate include microbial biomass
122 production, conversion to CH₄, trapping in reservoirs beneath impermeable caps, and
123 abiotic organic matter synthesis. Calcite deposition results in a temperature dependent
124 isotope fractionation and may be microbially mediated¹⁵⁻¹⁶ in cation-rich alkaline waters
125 with high aqueous CO₂ concentrations. Additionally, in the absence of light for
126 photosynthesis, microorganisms in subsurface settings instead fix CO₂ into biomass
127 through chemolithoautotrophy¹⁷⁻¹⁹. Both calcite deposition and chemolithoautotrophy
128 occur mostly at temperatures below 100°C, which are the prevalent conditions in the
129 forearc subsurface²⁰. Biologically induced carbon fixation generally results in a kinetic
130 fractionation of carbon isotopes that preferentially removes ¹²C from CO₂²¹. The degree of
131 fractionation depends on the microorganism's physiology, as well as substrate limitations,
132 temperature, and pressure. Biological activity can also decrease the carbon isotopic
133 composition ($\delta^{13}\text{C}$) of CO₂, by adding ¹³C-depleted carbon through heterotrophic
134 respiration of organic matter or methane. This process is not associated with a large
135 carbon isotope fractionation²¹ relative to pure kinetic reactions driven solely by biological
136 processes. In addition, abiotic hydrocarbon production from dissolved CO₂ has been
137 demonstrated in laboratory experiments over 250°C²², a possible temperature regime in
138 hotter forearc regions.

139

140 **He and C Results:**

141 We collected samples from 23 sites in northern and central Costa Rica (Figure 1),
142 which traversed forearc (Nicoya Peninsula), arc and backarc regions. Springs likely to
143 represent deeply-sourced fluids (based on field data such as temperature, dissolved
144 oxygen, salinity) were targeted, and samples were taken by inserting tubes into outflow
145 vents to minimize fluid interaction with atmosphere and surface water. The sites were
146 classified into two groups: the northern transect samples, which were collected to the
147 north of the EPR-CNS on-land extension, and the Central transect samples, which were
148 collected to the south of this plate transition. The two groups, with the exception of
149 Santa Lucia, are separated by the on-land extension of a major transition in the
150 downgoing slab: the boundary between oceanic crust formed at the East Pacific Rise
151 (EPR) and the Cocos Nazca Spreading (CNS) zone. We measured $^3\text{He}/^4\text{He}$ vs. air,
152 expressed as (R/R_A) , where $R = ^3\text{He}/^4\text{He}_{\text{sample}}$ and $R_A = ^3\text{He}/^4\text{He}_{\text{air}} = 1.39 \times 10^{-6}$ for 17
153 geothermal fluid (i.e., spring water) samples and 17 free gas samples. In addition, we
154 measured $\delta^{13}\text{C}$ vs. Vienna Pee Dee Belemnite (VPDB) in dissolved inorganic carbon (DIC)
155 in 31 geothermal fluid samples, dissolved organic carbon (DOC) in 16 fluid samples, and
156 CO_2 in 16 free gas samples (Supplementary Table 1). Sediments surrounding the surface
157 emanation of the springs were also sampled and total organic carbon (TOC) contents
158 and $\delta^{13}\text{C}$ were measured. The relative abundances of He to CO_2 are also reported
159 ($\text{CO}_2/^3\text{He}$; Extended Data Figure 1). The air-normalized $^4\text{He}/^{20}\text{Ne}$ ratio is used to
160 calculate the X-value, (multiplied by the Bunsen solubility (1.23) at the assumed
161 recharge temperature of 15°C^{23} for fluid samples), which is used to estimate the air-
162 corrected $^3\text{He}/^4\text{He}$ ratio (R_C) of the sample²⁴. As X-values are high (>5) for the majority of
163 samples of this study (Extended Data Figure 2), the correction factor is small and hence
164 there is typically little difference between measured (R/R_A) and corrected (R_C/R_A)
165 $^3\text{He}/^4\text{He}$ ratios (Supplementary Table 1).

166

167 The air-corrected $^3\text{He}/^4\text{He}$ (R_C/R_A) values range from 0.49 to 6.79 R_A
168 (Supplementary Table 1), with the highest values occurring in the volcanic arc and the
169 lowest occurring in the outer forearc. Arc values approach upper mantle values ($\sim 8 R_A$),

170 however, they exhibit evidence for a small but discernible addition of radiogenic helium
171 at all locations. In contrast, outer forearc and forearc values are predominantly
172 radiogenic, with a small but distinct mantle contribution, suggesting these fluids are an
173 admixture of crustal fluids and deeply sourced volatiles likely with some influence of
174 shallow water-rock interaction. He-isotope values are systematically higher (i.e., more
175 mantle-like) in both fluids and gases close to the volcanic arc-front and lower toward the
176 trench (i.e., in the forearc; Figure 2A), showing that slab/mantle outgassing is not limited
177 to the arc-front, and occurs throughout the entire forearc. However, free CO₂ gas
178 (bubbling springs) was only detected within ~20km of the arc (Figure 2B). In the absence
179 of gas manifestations, we examined evidence for forearc carbon processing in dissolved
180 carbon from spring outflows.

181

182 The $\delta^{13}\text{C}$ and carbon content of DIC and DOC within spring fluids decrease
183 systematically trenchward across the entire arc (Figure 2B and Figure 3). Strikingly,
184 DIC values in northern and central Costa Rica follow distinct parallel trends (Figure 3),
185 which are not masked by site-dependent surface-derived organic material
186 heterogeneities, such as vegetation, degradation of photosynthetic organic matter
187 and/or surface water infiltration, indicating that the distribution of carbon in these
188 fluids results from deep subsurface phenomena. In contrast, TOC is heavily
189 influenced by surface detrital carbon and shows no apparent trend between
190 concentration and $\delta^{13}\text{C}$ in samples across the two transects. DIC and DOC define
191 Rayleigh distillation curves, with northern springs showing consistently higher $\delta^{13}\text{C}$
192 for both DIC and DOC at a given DIC content. The parallel trends of decreasing $\delta^{13}\text{C}$
193 with decreasing DIC are consistent with isothermal (~65°C) precipitation of
194 isotopically heavy calcite from forearc fluids (see methods for details), with generally
195 higher degrees of calcite precipitation observed closer to the trench. This model is
196 supported by water chemistry data; PhreeqC was used to show that all outer forearc
197 samples are strongly saturated with respect to calcite and thus would be predicted to
198 readily precipitate calcite (Supplementary Table 3). We present a model for calcite

199 precipitation that closely fits the observed data distribution (Figure 3), but requires a
200 more positive $\delta^{13}\text{C}$ value of the deep total dissolved carbon (DC = DIC+DOC) source
201 fluid (+5.0‰) released from the slab in northern Costa Rica than in central Costa Rica
202 (+0.5‰). Volcanic gases, which are presumed to represent the primary magmatic
203 fluid, show isotopically more negative values than dissolved carbon in the calculated
204 DIC endmember fluids, due to the additional isotopic fractionation associated with
205 dissolution of CO_2 gas into an aqueous fluid²⁵ (Figure 3). Importantly, the modeled
206 initial dissolved carbon values thus reflect both the isotopic composition of the
207 subduction fluids and a fractionation effect derived from CO_2 dissolution from source
208 gases²⁶⁻²⁷ (Figure 3). Our data suggest that surface carbon expressions across the
209 forearc are ultimately inherited from slab/mantle fluids, which are intricately linked
210 to the tectonics of the region, and modified by broadly coherent shallow processes.

211

212 **Origin of fluids:**

213 The observation that northern and central Costa Rica samples have markedly
214 different source fluid compositions suggests a relationship to the character of the
215 subducting slab. The northern EPR crust has a higher density of seamounts and is
216 more permeable than the CNS crust²⁸. The higher resulting fluid flow through sub-
217 seafloor basalts at the EPR cools, hydrates, and chemically weathers the plate²⁹.
218 Building on previous models³⁰, we hypothesize that the higher fluid flow from this
219 water-rich downgoing slab flushes more carbon from the slab upward into the
220 overriding plate at shallower depths beneath the forearc, accounting for the greater
221 contributions of relatively ^{13}C -enriched carbonates in our northern sample set.
222 Additionally, enhanced fluid circulation in the EPR crust likely promotes calcite
223 veining in the downgoing plate, potentially providing an additional source of heavy
224 carbon³¹ in northern Costa Rica. Thus, the composition of the subducting slab is
225 expressed in surface-manifestations across the forearc. Our model predicts that
226 source fluids from northern Costa Rica are more ^{13}C -enriched than in central Costa
227 Rica (Figure 3). Both volcanic arc CO_2 and dissolved forearc carbon reflect a larger

228 carbonate contribution in northern Costa Rica, which is consistent with previous
229 observations in the volcanic arc which show an increased carbonate signal in
230 northern Costa Rica, which extends into Nicaragua^{30, 32-36}.

231

232 Importantly, the carbon isotope compositions of forearc springs can be used to
233 calculate the fraction of carbon lost through calcite deposition (see Figure 3 and
234 methods for details). Using the average $\delta^{13}\text{C}$ of the DC ($\delta^{13}\text{C}_{\text{DC}}$) of all the forearc springs
235 for each transect (northern = -8.8‰, central = -14.3‰) we find that 88% and 89% of the
236 total carbon released in the forearc is precipitated as calcite, respectively. Using the
237 average $\delta^{13}\text{C}_{\text{DC}}$ of the outer forearc springs (i.e., Nicoya Peninsula only; northern = -
238 14.4‰, central = -16.5‰) yields 95% and 92% of the total carbon precipitated as calcite,
239 for the northern and central transects, respectively. The close agreement between the
240 calculated fractions of total carbon lost to calcite is due to the fact that the Rayleigh
241 fractionation curves are steep at low $\delta^{13}\text{C}$ values (Figure 3). Thus, a large range in $\delta^{13}\text{C}$
242 observed in the forearc corresponds to a relatively narrow range of calculated
243 fractionation (F) values (i.e., fraction of carbon remaining; Figure 3). We conclude that
244 $91 \pm 4\%$ of forearc carbon is lost through calcite precipitation.

245

246 Measured DOC $\delta^{13}\text{C}$ values varied widely from -0.65 to -25.48‰ vs. VPDB,
247 with a mean value of -12.00‰. Like DIC, the lowest values were observed in the
248 outer forearc and the highest values were observed in the forearc, where values are
249 highly variable (Figure 2B and 2C). However, with the exception of one site in the
250 central region (Santa Lucia), DOC carbon isotope compositions and concentrations
251 strongly correlate with those of DIC (Figure 3), suggesting that DOC is produced from
252 deeply sourced fluids. Considering this, we propose a two-stage model whereby $91 \pm$
253 4% of the carbon released from the slab/mantle is consumed by calcite precipitation
254 and the remaining dissolved carbon is further fractionated by biological consumption.
255 Our model assumes a steady-state flux of deep (slab/mantle) fluids, which imparts
256 the carbon isotope signature of the slab into the upper plate where carbon is

257 partitioned into CO₂ gas, calcite, DIC, and DOC. DOC is potentially a mixture of deep
258 thermogenic DOC, deep microbial chemolithoautotrophy, and shallow
259 photosynthetic DOC. The outlier DOC sample, Santa Lucia, is likely dominated by
260 deep thermogenic DOC, due to the fact that it has an anomalously high DOC
261 concentration (5.69mmolC/L), $\delta^{13}\text{C}$ value (-0.65‰), and polycyclic aromatic
262 hydrocarbon concentrations (Supplementary Table 2), accompanied by a pronounced
263 visible oily sheen on the surface fluids. In all other sampling sites, DOC is likely
264 synthesized from DIC at physiologically feasible temperatures in the subsurface
265 rather than being a mixture of exogenous sources, since its concentrations and $\delta^{13}\text{C}$
266 values correlate tightly with DIC.

267

268 DIC and DOC carbon isotopes are offset from each other by $10.9 \pm 1.6\text{‰}$ in
269 the north, and $5.8 \pm 2.2\text{‰}$ in the central region (Figure 3); these $\Delta^{13}\text{C}_{\text{DIC-DOC}}$ values are
270 within the expected range for the reverse tricarboxylic cycle, which is a common
271 microbial carbon fixation pathway in the subsurface^{17,37}. The $\Delta^{13}\text{C}_{\text{DIC-DOC}}$ values are
272 also consistent with isotopic fractionation of hydrocarbons synthesized abiotically
273 from DIC at 250°C²². However, calcite precipitation occurs at ~65°C, and DOC
274 synthesis must occur after calcite precipitation for DOC carbon isotope compositions
275 to correlate with the post-precipitation DIC values. Therefore most of the DOC
276 appears to be derived from subsurface chemolithoautotrophy at <65°C. This is
277 further supported by the detection of microbial cells in all hot spring fluids except Las
278 Hornillas. We conclude that DOC at the remaining sites is primarily synthesized from
279 chemolithoautotrophy within the deep subsurface environment and that
280 slab/mantle-derived carbon is used by microorganisms to build biomass. By using cell
281 abundance values for the sampled subsurface fluids, we estimate that up to 2.8×10^9
282 moles of additional carbon could be locked into biomass in the Costa Rican forearc,
283 potentially contributing significantly to the overall carbon sink.

284

285 These findings have major implications for the global carbon mass balance of
286 subduction zones. We calculate a CO₂ flux from the forearc (Supplementary Table 4) by
287 combining the average DIC concentration in forearc waters of 4.24 mmol/L CO₂ with
288 measured flow rates, which ranged between 1-10 L/s and an estimated 50-500 springs³⁸
289 throughout the forearc region. We further consider that 5-50 of these springs have a
290 separate gas phase, with measured CO₂ fluxes of 201 mol × m⁻² × d⁻¹ to 655 mol × m⁻² ×
291 d⁻¹, and bubbling areas that typically cover 1 m² to 10 m² based on our field
292 observations. Using these broad criteria, the steady-state CO₂ flux is constrained to 7.1 ×
293 10⁶ to 7.9 × 10⁸ mol/yr (mean = 4.0 × 10⁸), which is two orders-of-magnitude smaller
294 than the volcanic-arc flux (1.3-6.1 × 10¹⁰ mol/yr)^{2,8}. As argued above, carbon isotopes
295 indicate that forearc calcite deposition and chemolithoautotrophy together remove
296 ~94% of the total carbon inputs. This increases the flux range that we calculate to 1.2 ×
297 10⁸ to 1.3 × 10¹⁰ mol/yr, which is significantly larger than the previous outer forearc
298 estimate of 2.1 × 10⁷ mol/yr, determined using a smaller number of submarine seeps³.
299 This represents up to 36% of the total volcanic-arc flux (= 3.7 × 10¹⁰ ± 66% mol/yr; taken
300 as the mean of previous CO₂ flux estimates from references^{2,8}; Figure 4; Supplementary
301 Table 4), which would result in a ~19% reduction in the amount of total carbon
302 transported to the deep mantle. Similarly, dissolved CH₄ concentrations and CH₄/CO₂ in
303 sampled fluids are used to constrain the CH₄ flux from 5.1 × 10³ to 1.0 × 10⁶ mol/yr.
304 Thus, CH₄ emissions contribute negligibly to the total forearc C budget.

305

306 **Implications for evolution of Earth Reservoirs:**

307 Our results suggest that significantly less carbon is transported into Earth's
308 mantle than previous estimates suggest¹, due to the previously unrecognized calcite
309 forearc carbon sink. This observation has wide ranging implications for the volatile
310 inventory of the mantle and the temporal evolution of Earth reservoirs, which controls
311 the planet's redox balance³⁹⁻⁴¹. For example, the nature (reduced vs. oxidized carbon)
312 and extent of carbon reaching the mantle is intricately linked to volatile recycling, and
313 impacts the net oxidation state of Earth's surface and deep interior. Furthermore, we

314 propose that carbon sequestration in the forearc may be directly related to plate
315 subduction angle, which dictates the extent of forearc. Globally, flat subduction zones
316 represent only about 10% of total subduction zones⁴², which would indicate that
317 approximately 2% less carbon than previously estimated is currently delivered to the
318 deep mantle. However, such a carbon sink may have been enhanced during the late
319 Archean, when oceanic crust was subducted at an anomalously low-angle⁴³. Notably,
320 evidence for the so-called Archean flat-subduction is preserved in most late Archean
321 (3.0–2.5 Ga) terrains, but is lacking in the early Archean (>3.3 Ga). If carbon was stored
322 overwhelmingly on continental (or island arc) margins, then continental collisions and
323 continental arc flare-ups would be the most important regulator of atmospheric CO₂
324 through time. During the Proterozoic, subduction zones were dominated by high dip
325 angles and smaller forearcs⁴⁴, enabling volatiles to be more efficiently transferred into
326 deep mantle, as the forearc carbon sink would likely be less significant and transfer to
327 the mantle would be more efficient. A more efficient transfer of reduced carbon to the
328 mantle could then lead to enhanced accumulation of oxygen in the atmosphere over
329 time. Thus, this work provides potential forcing mechanisms on the great oxygenation
330 event, which was certainly controlled in part by subduction efficiency and plate
331 tectonics⁴⁵⁻⁴⁷. In addition, these findings also have implications for the modern
332 atmosphere, as better constraints on the long-term volcanic carbon budget and how it
333 may be buffered by chemical and biological processes is critical for evaluating natural
334 and anthropogenic climate forcings.

335

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577 P.H.B. originally conceived the idea for the project, was lead-PI on the Sloan (Deep
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588 provided comments on the manuscript and either assisted in sample collection (as
589 part of the Biology Meets Subduction team or on previous expeditions) and/or
590 analyzed samples in their respective laboratories.

591

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593 The authors declare no competing interests. Reprints and permissions information is
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596

597 **Figure 1** – Map of the northwest coast of Costa Rica. Northern sites (blue) and central
598 sites (orange) as well as the plate boundary between the East Pacific Rise (EPR) crust
599 and Cocos Nazca spreading center (CNS) crust, and the Quesada Sharp Contortion (QSC)
600 are shown. Sample distances to trench (Supplementary Table 1) were calculated as the
601 distance of a line segment extending from the sample location, along the angle of

602 convergence, to the plate boundary⁴⁸. Red-tipped triangles are volcanoes Orosí, Rincón
603 dela Vieja, Miravalles, Tenorio, Arenal, Platanar, Poás, Barva, Turrialba, and Irazú, from
604 north to south. Thin lines are depth to slab in 20km intervals⁴⁹. Background data from
605 GeoMapApp (<http://www.geomapapp.org>)⁵⁰.

606

607 **Figure 2A** – Helium isotopes ($^3\text{He}/^4\text{He}$) vs. distance from trench. The plot shows a
608 decrease in $^3\text{He}/^4\text{He}$ trenchward, suggesting that there is pervasive deep slab/mantle
609 degassing occurring across the entire arc, even though free CO_2 gas is absent. **2B** –
610 carbon concentrations (as indicated by circle size) and $\delta^{13}\text{C}$ of CO_2 gas, dissolved
611 inorganic carbon (DIC), dissolved organic carbon (DOC), and total organic carbon in
612 sediments surrounding the surface emanation of the springs (sTOC) are shown vs.
613 distance from trench and subduction zone section (**C**) in order to show the full range
614 in data. These “bubble plots” show that there is a corresponding decrease in carbon
615 concentrations and $\delta^{13}\text{C}$ values of DIC and DOC trenchward, and no change in
616 concentrations and $\delta^{13}\text{C}$ values of TOC.

617

618 **Figure 3** – Carbon isotopes as a function of DIC concentrations for northern (blue)
619 and central (orange) DIC (filled symbols), along with isotope fractionation model in
620 solid lines. DOC $\delta^{13}\text{C}$ values (open markers) as well as DOC concentrations (inset)
621 correlate with those of DIC. For clarity, Santa Lucia (5.69 mmol/L DOC) is not included
622 in inset. Values for arc gases ($T \geq$ boiling; plotted at 0% C sequestered as calcite)
623 represent an endpoint of CO_2 concentration and $\delta^{13}\text{C}$, offset from DIC by the
624 fractionation from gas to aqueous phase.

625

626 **Figure 4A**– Box model showing the carbon inputs⁸ in the Costa Rica Forearc.
627 Estimated inputs are from sediments, altered oceanic crust and serpentinized mantle
628 via subduction. Estimated outputs are shown for the forearc (this study) and the arc^{2,8},
629 as well as predicted transport to the deep mantle (inputs minus outputs). **4B** – Inset
630 box model showing endmember carbon isotopic values into the forearc following

631 release from the slab/mantle. The model shows fluid pathways and associated
632 fractionation as calcite forms, with approximately 91% of total carbon released in the
633 forearc sequestered as calcite. The remaining dissolved carbon (DC) is isotopically
634 depleted and partitioned into dissolved inorganic carbon (DIC) and dissolved organic
635 carbon (DOC).

636

637 **Methods:**

638 **Sample collection and analysis:**

639 In 2017, fluid and gas samples were collected across two transects of the
640 Costa Rican Arc (Figure 1), with pH ranging from 0.9-10.0 and temperatures ranging
641 from 23-89°C. Fluid and gas samples were collected in evacuated glass flasks and Cu-
642 tubes using standard collection procedures⁵¹, whereby precautions were taken to
643 minimize any possible air contamination²⁷. Gas and fluids samples (n=24) from 2017
644 are accompanied by addition (n=17) samples collected during reconnaissance field
645 campaigns in 2008, 2010 and 2012. He and C isotope data from the initial
646 reconnaissance field campaigns were produced in the Fluids and Volatiles Laboratory
647 at Scripps Institution of Oceanography (SIO), whereas data from 2017 are from
648 Oxford (He-isotopes) and Tokyo Institute for Technology (C-isotopes).

649

650 Samples were collected over several field campaigns; however, all samples
651 were collected during the dry season in an effort to minimize seasonal effects
652 (Supplementary Table 1). Considering this, the data from the various labs are in good
653 agreement. For example, fluids were collected at the Sabana Grande site in 2008,
654 2010 and again in 2017; C-isotope values of -12.69‰ (2017; Japan) agree well with -
655 12.75‰ (2008; SIO), -12.79‰ (2008; SIO) and -13.43‰ (2012; SIO). He-isotope
656 values for this site were $2.66 \pm 0.13 R_A$ (2017; Oxford), $0.60 \pm 0.03 R_A$ (2008; SIO) and
657 $1.04 \pm 0.11 R_A$ (2012: SIO), all indicate an admixture of radiogenic and mantle derived
658 gases. Notably, there is not thought to be a discernable cosmogenic component in
659 subduction-related fluids, as ³He outputs greatly exceed input parameters⁶. Also,

660 much too little interplanetary dust particles is available to sustain the flux required.
661 See Trull 1994. Also, high diffusivity of He in most low-T minerals is important. When
662 considering the reproducibility of He isotope measurements, it is essential to
663 consider the amount of air contamination in a given sample, which is estimated using
664 the relative amount of He and Ne, expressed as the X-value ($^4\text{He}/^{20}\text{Ne}$ normalized to
665 air). At Sabana Grande, the most pristine sample (highest X-value) yielded the highest
666 He isotope value of 2.66 R_A (Extended Data Figure 2). Helium isotopes were also
667 measured in samples collected at the Pueblo Antiguo site in 2010 (SIO), 2012 (SIO)
668 and 2017 (Oxford), and in the two samples with high X-values (>5), the He isotopes
669 measured at different laboratories are within analytical error (Oxford, 2017 = $4.34 \pm$
670 $0.22 R_A$ and SIO, 2010 = $4.51 \pm 0.11 R_A$).

671

672 Gas and water samples from the 2008, 2010 and 2012 campaigns were analyzed
673 at SIO for helium and carbon isotopes using instrumentation and protocols described
674 previously^{2,7,52}. All samples were extracted on a dedicated preparation line with a
675 fraction of the non-condensable gas (containing He and Ne) captured in a 1720-glass
676 breakseal. All CO_2 was condensed into a Pyrex breakseal following separation from
677 water vapor, non-inert gases (N_2 , CO , CH_4) and heavy noble gases (Ar, Kr and Xe).

678

679 Helium and neon analyses were carried out on a MAP-215 noble gas mass
680 spectrometer SIO. First, the gas was released from the breakseal and prepared for
681 analysis using a series of traps, held at liquid nitrogen temperature, and active-gas
682 getters. Helium was separated from neon using a helium-cooled refrigeration stage
683 interfaced to a trap lined with activated charcoal. All sample $^3\text{He}/^4\text{He}$ ratios were
684 normalized to standard aliquots of air processed and analyzed under identical
685 conditions.

686

687 Carbon dioxide was purified at SIO on a dedicated line using a variable
688 temperature trap designed to separate CO_2 from sulfur-bearing species. Following

689 cleanup, the amount of CO₂ was measured using a capacitance gauge manometer in a
690 calibrated volume, prior to freezing an aliquot of the CO₂ in a Pyrex breakseal. For
691 isotope analysis, the CO₂ aliquot was inlet into a Thermo-Finnigan Delta XP^{Plus} Isotope
692 Ratio Mass Spectrometer (IRMS). Carbon isotopes on gas samples from the 2017
693 campaign were analyzed at Universidad Nacional on a Picarro G2201-I by acidification of
694 NaOH solutions extracted from Giggenbach bottle samples. $\delta^{13}\text{C}_{\text{PDB}}$ values were
695 calibrated against a set of 8 standards with values ranging from + 2.42 ‰ to -37.21‰,
696 including internationally accepted standards NBS19 and Carrara Marble. Reported delta
697 values have uncertainties of <0.1 ‰ based on repeat analyses of standards and samples.
698

699 Noble gas analysis was also conducted in the Noble Laboratory at the University
700 of Oxford (2017 samples), using a dual mass spectrometer setup, interfaced to a
701 dedicated extraction and purification system⁵³. Gases were collected in Cu-tubes, and
702 then transferred to the extraction and purification line where reactive gases were
703 removed by exposing gases to a titanium sponge held at 950 °C. The titanium sponge
704 was cooled for 15 minutes to room temperature before gases were expanded to a dual
705 hot (SAES GP-50) and cold (SAES NP-10) getter system, held at 250 °C and room
706 temperature, respectively. A small aliquot of gases was segregated for preliminary
707 analysis on a Hiden Analytical HAL-200 quadrupole mass spectrometer. All noble gases
708 were then concentrated using a series of cryogenic traps; heavy noble gases (Ar-Kr-Xe)
709 were frozen down at 15 K on an all SS finger and the He and Ne were frozen down at 19
710 K on a cold finger filled with charcoal. The temperature on the charcoal finger was then
711 raised to 34 K to release only He, which was inlet into a Helix SFT mass spectrometer.
712 Following He analysis, the temperature on the charcoal cryogenic trap was raised to 90
713 K to release Ne, which was inlet into an ARGUS VI mass spectrometer.

714

715 Water samples for carbon isotope analysis (2017 samples) of dissolved inorganic
716 carbon (DIC), dissolved organic carbon (DOC) were collected by 50-mL syringes and
717 filtered by connecting membrane syringe filters with a pore size of 0.20 µm (DISMIC–

718 25AS; Advantec Toyo Kaisha, Tokyo, Japan) and syringe needle. Then filtered water was
719 directly injected to the pre-vacuumed 50-mL serum bottle sealed with butyl rubber
720 septa and an aluminum crimp. The subsampled water (10 mL) for DIC measurement
721 from each water sample of 50-mL sealed vials by nitrogen gas and transferred to a pre-
722 vacuumed 30-mL glass vial sealed with butyl rubber septa and an aluminum crimp. DIC
723 concentrations and their $\delta^{13}\text{C}$ values were measured using CO_2 in the headspace of glass
724 vials after a 1-h reaction with injected 0.5mL H_3PO_4 . DOC were also measured CO_2 in the
725 headspace after the reaction of carbonate-free residue with 0.2 g sodium persulfate.
726 The amount of CO_2 and the isotopic values were measured using a Agilent 6890N gas
727 chromatograph attached to a Thermo-Finnigan Delta XP^{plus}. Two international standards
728 ($\delta^{13}\text{C} = -13.90\text{‰}$ and 2.52‰) were used for standardization, and the standard
729 deviations were shown by over 3 times measurement.

730

731 The sediments surrounding the surface emanations of the springs were collected
732 for measuring TOC content and carbon isotopic compositions. The sediment samples
733 were kept at 4°C until transporting to the laboratory. Glass vial samples were then
734 stored at -80°C until further treatment. Firstly, the sediment samples were freeze-dried
735 and then crushed into fine grains using a mortar to remove large leaves and roots. 50–
736 100 mg of sediment samples were weighed and reacted with 1 M HCl solution until
737 effervescence stopped, followed by a rinse with distilled water until the pH neutralized.
738 The TOC sediment samples were analyzed using an elemental analyzer (EA-1110;
739 Thermo Fisher Scientific) coupled to a Thermo Fisher Scientific MAT 252 isotope ratio
740 mass spectrometer (IRMS).

741

742 Samples for cell counts were taken in 2017 as close to the source spring as
743 possible, usually in an outflow from a rock outcrop or a small surface pool that was
744 rapidly being refilled by the source. We placed 1 ml fluids into a 2 ml plastic tube with a
745 rubber o-ring screwcap (to prevent evaporation) containing 500 μl 3%
746 paraformaldehyde solution in phosphate-buffered-saline (PBS). Cell count samples fixed

747 with 3% paraformaldehyde in the field were kept at room temperature during return to
748 the University of Tennessee and were weighed upon returning to the lab. Cell counts
749 were determined on a Guava Easy Cyte 6HT-2L (Millipore) flow cytometer. Triplicate
750 aliquots of each sample (200 μ L) were stained with 5 \times SybrGreen prior to analysis. We
751 estimated contributions of cell biomass with total cell counts, and average carbon
752 content of subsurface microbes⁵⁴, by multiplying the average number of cells in our fluid
753 samples by the volume of hosting rocks (from the trench to the arc – assuming a log
754 increase of the isotherms moving toward the arc) up to 2 km depths, and using an
755 average rock porosity (to obtain possible fluid amounts)⁵⁵, and found that they did
756 represent a significantly large carbon reservoir.

757

758 Aliphatic hydrocarbons (C10-C40) and polycyclic aromatic hydrocarbons
759 (PAHs) were analyzed using conventional procedures based on gas chromatography
760 with a flame ionization detector (FID) and HPLC with diode array and fluorimetric
761 detection⁵⁶. Briefly, aliphatic hydrocarbons (C10-C40) were extracted with
762 hexane:acetone (2:1) in a microwave (110 $^{\circ}$ C for 25 min, 800 Watt) (Mars CEM, CEM
763 Corporation, Matthews NC). After centrifugation at 3.000 \times g for 10 min, the
764 supernatants were purified with solid-phase extraction (Phenomenex Strata-X, 500
765 mg \times 6 mL plus Phenomenex Strata-FL, 1000 mg \times 6 mL) and then concentrated using
766 a SpeedVac (RC1009; grade n-hexane and analyzed with a PerkinElmer gas
767 chromatograph) equipped with an Elite-5 capillary column (30 mm \times 0.32 mm ID \times
768 0.25 μ m-df) and a FID. For quantitative determination, the system was calibrated
769 with an unsaturated pair n-alkane standard mixture according to ENISO 9377-3 (Fluka
770 68281). For the analysis of PAHs, sediment samples were extracted using 0.5 M
771 potassium hydroxide in methanol with microwave at 55 $^{\circ}$ C for 20 min (800 Watt)
772 (CEM, Mars System). After centrifugation at 3.000 \times g for 10 min, the methanol
773 extracts were concentrated using a SpeedVac and purified with solid-phase
774 extraction (Octadecyl C18, 500 mg \times 6 mL, Bakerbond). A final volume of 1 mL was
775 recovered with pure, analytical HPLC gradient grade acetonitrile; HPLC analyses were

776 carried out in a water-acetonitrile gradient by fluorimetric and diode array detection.
777 The PAHs were identified according to the retention times of an appropriate pure
778 standards solution (EPA 610 Polynuclear Aromatic Hydrocarbons Mix), and classified
779 as low molecular weight (LMW: naphthalene, acenaphthylene, 1-methyl
780 naphthalene, 2-methyl naphthalene, acenaphthene, fluorene, phenanthrene,
781 anthracene) or high molecular weight (HMW: fluoranthene, pyrene,
782 benzo(a)anthracene, chrysene, 7,12-dimethyl-benzo(a)anthracene,
783 benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene,
784 dibenzo(a,h)anthracene, benzo(g,h,i)perylene, indeno(1,2,3-cd)pyrene). Accuracy
785 and precision were checked analyzing both pure standard solutions and reference
786 materials (NIST 1944) and the obtained concentrations were always within the 95%
787 confidence intervals of certified values. Aliquots of all the samples were dried in a
788 oven at 60°C for at least 8 h, up to obtain a constant weight, in order to quantify the
789 interstitial water content, allowing to express all the analyzed chemicals as a function
790 of the dry weight (d.w.) of the sediments.

791

792 **Isotope fractionation modeling:**

793 He-C studies in volcanic arc settings^{2,57-60} have coupled helium and carbon
794 isotopes to distinguish carbon from different provenances using a three-component
795 mixing model (Extended Data Figure 1). In such acidic settings, volcanic activity has been
796 suggested to release previously sequestered CO₂⁶¹⁻⁶⁴, which mixes with slab/mantle
797 carbon and results in the characteristic signatures. Samples from this study were
798 collected from a range of lithologies from ophiolites in Nicoya⁶⁵⁻⁷⁰ to andesitic and
799 basaltic volcanic rocks in the arc and forearc. The volcanic front in Costa Rica developed
800 on the western edge of the Caribbean Plateau, an oceanic plate basement without any
801 preexisting continental or arc material⁷¹⁻⁷³. The Santa Elena ophiolite is part of a series
802 of exotic, arc-related terranes that make the basement of the volcanic front in southern
803 Nicaragua and does not extend south of the Hess Escarpment-Santa Elena suture⁷⁴⁻⁷⁵.
804 However, these carbon isotope data are consistent with a model that requires for a

805 homogenous C isotope input flux. This indicates that degassing of the downgoing
806 slab/mantle drives the ^{13}C -isotope inputs, rather than the over-riding crust, but in some
807 cases, this signal is subsequently obfuscated by low temperature precipitation of
808 isotopically heavy calcite, which effectively fractionates the carbon isotopes. Here we
809 provide additional details on the fractionation modeling used to construct the calcite
810 fractionation curves in Figure 3 (main text).

811 In summary, CO_2 is released from the slab/mantle and reacts with shallow
812 groundwater forming an initial DIC pool (dissolved CO_2). A fractionation factor (see
813 details below) is calculated based on temperature (assumed to be 67°C in the northern
814 transect and 63°C in the central transect) and an alpha value for $\text{CO}_2\text{-HCO}_3^-\text{-CaCO}_3$,
815 where CaCO_3 is calcite. The starting $\delta^{13}\text{C}$ input is determined to be different from the
816 northern (5.0‰) and central (0.5‰) transect, presumably controlled by different slab
817 inputs. Notably, the presumed slab inputs are consistent with positive isotope values
818 measured in carbonate sediments off the coast of Nicoya⁷⁶. Starting $\delta^{13}\text{C}$ input
819 conditions are assumed to be the same for forearc, backarc and arc. DOC is in isotopic
820 equilibrium with DIC at these temperatures and assumed alpha values. The DIC is
821 transformed from CO_2 to HCO_3^- in alkaline waters and the resulting DIC concentrations
822 are not affected by further dilution of meteoric water or by concentration due to steam
823 loss. As the Rayleigh fractionation progresses, the DIC in the forearc is consumed and a
824 Rayleigh fractionation occurs. Open system fractionation (Rayleigh distillation) curves
825 were iteratively fitted to the data using:

826

827 eq. 1.

$$828 \delta^{13}\text{C}_{\text{DC}f} \cong (\delta^{13}\text{C}_{\text{DC}i} + 1000)(F^{\alpha_{\text{DC-calcite}}-1}) - 1000$$

829

830 after⁷⁷, where F is the fraction of dissolved carbon (DC) remaining in the fluid, $\delta^{13}\text{C}_{\text{DC}f}$
831 is the carbon isotope composition of DC at F, $\delta^{13}\text{C}_{\text{DC}i}$ is the initial isotope composition
832 of the DC, and $\alpha_{\text{DC-calcite}}$ is the fractionation factor between DC and calcite at a given
833 temperature. The fractionation factor between DC and calcite was calculated from⁷⁸:

834

835 eq. 2.

836

$$837 \quad 1000 \ln \alpha_{DC-calcite} = -8.91 \times 10^8 T^{-3} + 8.557 \times 10^6 T^{-2} - 1.881 \times 10^4 T^{-1} + 8.27$$

838

839 where T is the temperature in Kelvin.

840

841 Model curves were fit to the data from each transect by iteratively varying the
842 values of $\delta^{13}C_{DC_i}$ and the temperature of calcite formation. Best fit results were
843 obtained with $\delta^{13}C_{DC_i} = +5.0\text{‰}$ and $T = 67 \text{ °C}$ ($R^2 = 0.91$) for the northern transect and
844 $\delta^{13}C_{DC_i} = +0.5\text{‰}$ and $T = 63 \text{ °C}$ ($R^2 = 0.86$) for the central transect.

845

846 Total dissolved carbon (DC) values used in the model are calculated from the sum of
847 measured DIC and DOC concentrations (Supplementary Table 4), and $\delta^{13}C_{DC}$ is the
848 average of $\delta^{13}C_{DIC}$ and $\delta^{13}C_{DOC}$, weighted by their respective DIC and DOC
849 concentrations. DIC dominates total DC contents, typically accounting for ~80% of DC
850 on a sample by sample basis. DOC data are not available for 12 of our 30 samples, as
851 several samples were collected on previous reconnaissance field campaigns.

852 However, the strong correlation between DC and DIC (Extended Data Figure 3) allows
853 us to confidently predict the DC concentration and $\delta^{13}C_{DC}$ of the samples for which
854 DOC is not available. The predicted DC and $\delta^{13}C_{DC}$ are thus used for the 12 samples
855 for which DOC data are not available in Figure 3 (main text). The slope of the linear
856 regressions in Figures S2a and S2c is used to calculate the fraction of DOC in the
857 northern and central transects (5.4 % and 4.7 %, respectively).

858

859 **Carbon budget calculations:**

860 The carbon isotope fractionation model allows calculation of the fraction of
861 carbon in various forearc reservoirs. Based on the average $\delta^{13}C_{DC}$ values at the forearc
862 sites (here defined as located at <155 km from the trench), the fraction (F) of carbon

863 remaining in the fluid can be calculated (expressed as "% carbon sequestered as calcite"
864 in Figure 3, which is equal to $100(1 - F)$). The average $\delta^{13}\text{C}_{\text{DC}}$ values at forearc sites are
865 further used to calculate the total DC concentration in mmol/L based on the Rayleigh
866 curves. Because the curves are steep at low F values, the $\delta^{13}\text{C}_{\text{DC}}$ values represent a more
867 robust and sensitive parameter than DC concentrations, which are more susceptible to
868 dilution and analytical uncertainty at low concentrations. The average DIC concentration
869 for forearc fluid is then calculated from dissolved carbon concentrations from the
870 relationship between DC and DIC expressed in Figures S2a and S2c. In this model, DOC
871 is then the residual carbon once DIC and calcite concentrations are accounted for. The
872 results of these calculations indicate that 91.3% of the total forearc carbon is
873 sequestered as calcite, 6.0% is released at the surface as DIC, and 2.7% is consumed by
874 microbiological processes to form DOC.

875

876 Our flux estimates for the Costa Rican forearc (see main text) show that between
877 7.1×10^6 and 7.9×10^8 mol CO_2/yr are released to the surface by spring outflow as DIC
878 and CO_2 gas. Based on the above-described model, we argue that this represents the
879 residual carbon left over after pervasive calcite deposition. The total flux (i.e., subducted
880 slab and mantle fluid) through the forearc, including both calcite deposition and
881 microbial consumption, ranges from 1.2×10^8 to 1.3×10^{10} mol/yr (Figure 4).

882

883 **Previous Mass Balance calculations:**

884 To put our forearc flux estimate into context, we must consider previous flux
885 estimates, which to date, have focused mostly on high temperature regions¹¹⁻¹³, where
886 there is a distinct point-source for carbon emissions. However, little work has been
887 conducted in the forearc of Costa Rica³ even though thermodynamic modeling of the
888 Costa Rica convergent margin predicts efficient CO_2 release from the top of the
889 subducting slab beneath forearcs, at depths of 65-80 km¹².

890

891 Past budget approaches⁵⁸ coupled He and C isotopes to tease apart C sourced
892 from different provenances using a three-component mixing model (Fig. S1). Subducted
893 slab carbon is presumed to derive from carbonate rocks and have a value of ~ 0 VPDB,
894 mantle values are assumed to be approximately $-6.5 \pm 2.0\%$ and isotopically low
895 carbon isotope values are attributed to contributions from organic sediments (-30%).
896 Here we argue that in the forearc environment these values do not result from mixing,
897 but instead from low temperature calcite precipitation and biological consumption.

898

899 Arc fluxes are typically only measured along a relatively narrow transect source:
900 the volcanic front. In contrast, springs occur over a large stretch of the forearc, so this is
901 a much larger area to integrate. Quantitative assessment of gas fluxes from the forearc,
902 backarc, fault-related sites, hydrothermal volcanic systems (e.g., Tenorio, Miravalles,
903 Irazú, Rincón de la Vieja flank sites), and concurrent diffuse degassing are lacking. Here
904 we place better constraints on forearc fluxes, which complement recent constraints on
905 magmatic C fluxes⁸. We estimate a flux between of 1.2×10^8 and 1.3×10^{10} to mol CO₂
906 per year, which represents as much as $\sim 36\%$ of the total arc flux. , and could reduce the
907 amount transferred into the deep mantle by up to $\sim 19\%$.

908

909 Previous studies have utilized helium and carbon isotopes ($^3\text{He}/^4\text{He}$, $\delta^{13}\text{C}$) and
910 relative abundances ratios (CO₂/ ^3He) of fumarole sites at arc settings. Two approaches
911 were taken in these areas in order to determine CO₂ fluxes: 1) combining measured
912 CO₂/ ^3He data with known ^3He fluxes and 2) to combine measured CO₂/SO₂ data with
913 known SO₂ fluxes. Previous work was conducted throughout Costa Rica^{3,6,8} El Salvador
914 Honduras⁷ and Nicaragua².

915

916 In typical subduction zones, the vast majority of carbon is thought to be recycled
917 back into the lithosphere, hydrosphere, and atmosphere¹, and what remains is
918 transported into the deep mantle. In stark contrast, mass balance considerations from
919 the Costa Rica convergent margin suggest the majority of subducted C (more than 85%)

920 is ultimately transferred into the deep mantle^{2,3,6}. Importantly, this estimation assumes
921 that the forearc and backarc fluxes are negligible, which we have shown in this
922 contribution to be untrue. Prior work⁶ estimated the subducting CO₂ input flux for ~310
923 km Arc of Costa Rica to be 8.2×10^{10} mol/yr⁶, assuming “1% organic CO₂” in sediments
924 based on data from a single IODP site (1039) and a “global average” altered crustal
925 composition of 0.2% CO₂, extrapolated over the entire slab thickness. Published carbon
926 degassing (output) flux estimates vary between 1.2×10^{10} mol/yr to 1.5×10^{10} mol/yr,
927 with an average value of 1.26×10^{10} mol/yr. Importantly, these estimates are
928 determined to be >90% sediment and marine carbonate derived, based on C-isotope
929 data². However, more recent studies^{8,61,79} use a more complete assessment of volcanic
930 degassing from the arc to show that the current volcanic CO₂ flux from Costa Rica is
931 significantly higher (6.10×10^{10} mol/yr). The 2017 study⁸ is based on new data for 11
932 volcanoes in Costa Rica and Nicaragua showing that the CO₂ flux from the arc is
933 underestimated in previous studies. Their finding suggests that the Central American
934 subduction margin may be more similar to other arc segments¹ than previously
935 thought^{2,6}. The 2017 findings⁸ suggest the 2014-2015 CO₂ flux from the Costa Rican arc
936 is an order of magnitude greater than previous estimates^{2,6}. However, the Costa Rican-
937 Nicaraguan margin has displayed an increase in arc-wide volcanic activity in recent
938 years⁸. Thus, arc systems probably show significant variation in gas output over time. If
939 the older estimates^{2,6} of CO₂ flux are taken to be more representative of the time-
940 integrated CO₂ flux, then there is still a considerable missing flux in the forearc or arc. In
941 Figure 4 we take the average value from references 2 and 8 to determine an average
942 CO₂ flux of $3.7 \text{ mol/yr} \pm 66\%$.

943

944 **Geologic Background:**

945 The Central American subduction zone is characterized by rapid (70–90 mm/year)
946 convergence of young (15–25 Ma) oceanic lithosphere⁸⁰⁻⁸², whereas the downgoing Cocos
947 plate is composed of a thin sedimentary layer (~428m). The upper section of the column
948 (177 m) consists of Quaternary to upper Miocene hemipelagic diatomaceous mud and

949 middle Miocene brown abyssal clay and the lower section consists of middle–lower
950 Miocene chalky carbonate ooze and manganiferous chalk and chert⁸³⁻⁸⁴. The composition
951 of subducted sediments and carboniferous material is microbiologically altered, due to
952 the fact that anaerobic respiration and fermentation determine sediment redox
953 conditions, alkalinity, and carbon isotope pools⁸⁵⁻⁸⁶. The entire column of incoming
954 sediments under-thrusts the toe of the Caribbean Plate. However, the absence of
955 significant off-scraping or sediment accretion suggests the Costa Rica convergent margin
956 is either non-accretionary, or underplated further landward^{79,87}. Under-thrusting leads to
957 rapid compaction of the upper hemipelagic sediments and dewatering of pore fluids
958 within the first few kilometers of subduction⁸⁸, resulting in the release of carbon dioxide
959 and methane at relatively shallow depths into the overlying forearc³. Fluids released
960 during dehydration reactions ascend through the overriding plate along deep-penetrating
961 faults, producing numerous fluid venting sites in the forearc of Costa Rica⁸⁹. A significant
962 portion of the forearc is subaerial (Nicoya and Osa peninsulas), providing unique sampling
963 opportunities not found at the majority of arc systems. In addition to mechanically
964 induced shallow dewatering near the toe of the overriding plate, deeply-sourced (i.e.,
965 abiotic) fluids are released by mineral dehydration reactions and phase transformations at
966 depths of approximately 10-15 km (between 60-140°C)^{88,90}, which are pressure and
967 temperature ranges that are within the range of theoretically habitable environments for
968 microorganisms⁹¹⁻⁹². Volcanoes in Costa Rica form a part of the southern segment of the
969 Central American Volcanic Arc. The volcanic arc in Costa Rica extends from the north to
970 the center of Costa Rica, immediately to the east of the Nicoya Peninsula. We collected
971 samples for this study in the forearc, arc and backarc of this region.

972

973 **Microbiological Background:**

974 ¹³C-depleted carbon dioxide in fluids released in the submarine portion of the
975 outer forearc indicate a $\delta^{13}\text{C}$ depleted source and may imply biological production
976 through anaerobic respiration of sulfate, which is an abundant oxidant in seawater
977 (Füri et al. 2010). The level of ¹³C-depletion can be moderate (-10 to -15%),

978 suggesting electron donors from organic matter, or extreme (-60 to -70‰),
979 suggesting electron donors from methane. Methanotrophy can occur anaerobically
980 through sulfate reduction⁹³, or aerobically when oxygenated seawater mixes with
981 methane-rich fluids⁹⁴. The methane itself may be methanogenic products mobilized
982 from the >12 km source fluids⁹⁵, or shallower methanogenesis⁹⁶, but this has yet to
983 be resolved with isotopic analysis.

984

985 Methane in the subaerial section of the margin has been found to have a
986 significant biogenic component, with $\delta^{13}\text{C}$ of -35 to -45‰, and presence of
987 methanogenic archaea⁹⁷⁻⁹⁸. However, no large-scale survey of the effects of
988 microbiology on degassing across a convergent margin have been performed. The
989 few microbiological studies that have been performed in convergent margins have
990 focused on a single site at a time, preventing any region-scale exploration of how
991 microbes interact with these deep geological processes.

992

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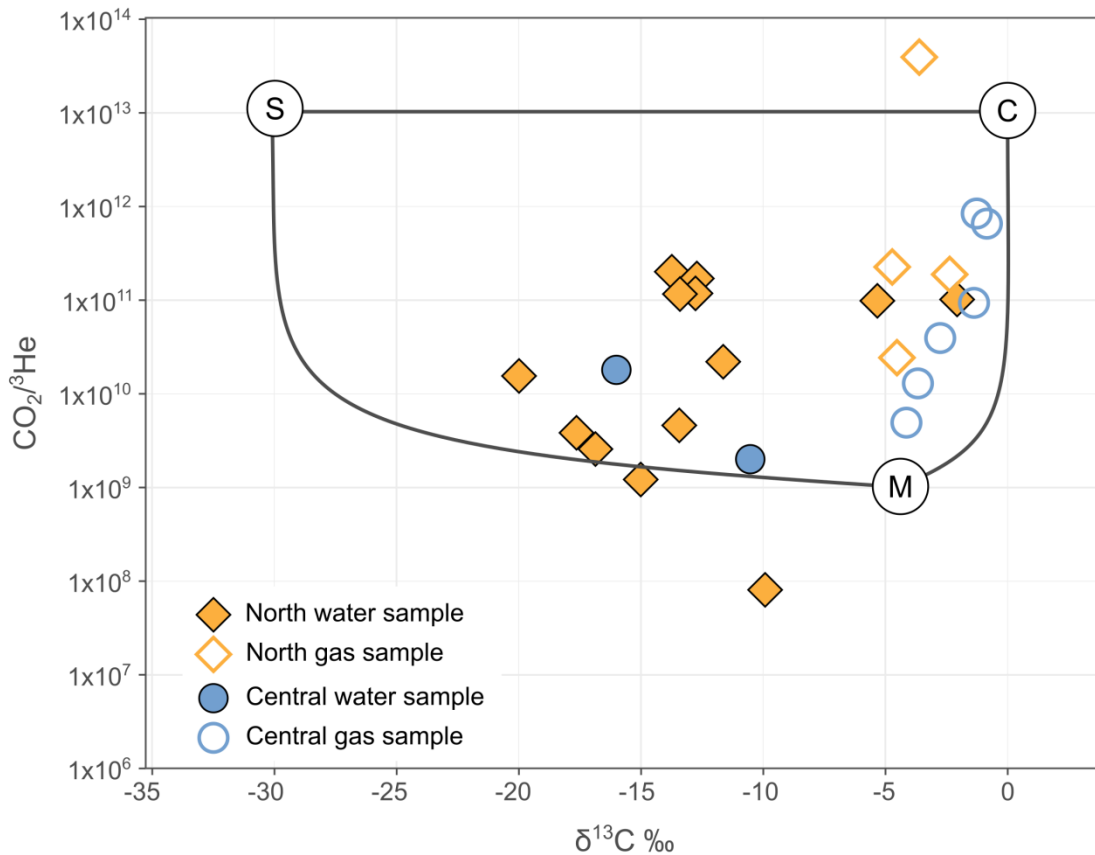
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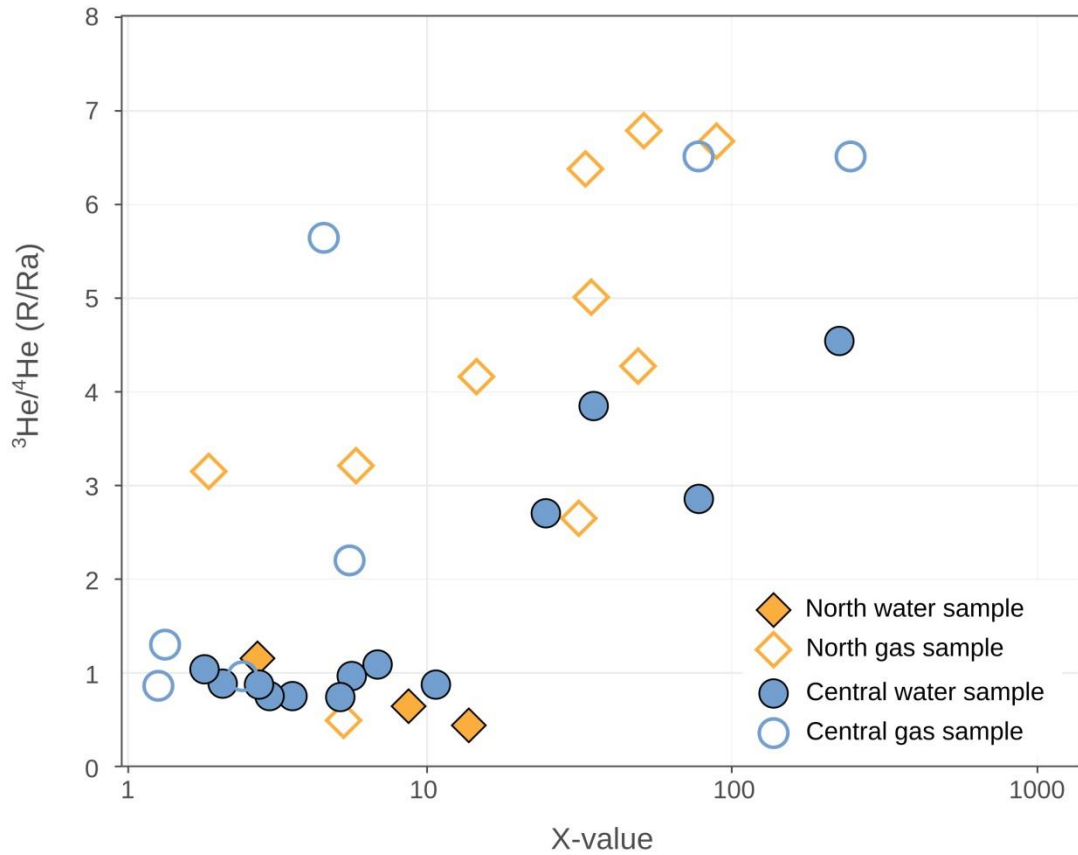
1209 **Data Availability Statement (DAS):**

1210 All raw data needed to make the plots are available in Tables 1 and 2 as well as in the
1211 Source Data file provided. All data is archived through EarthChem (DOI:
1212 10.1594/IEDA/111271)
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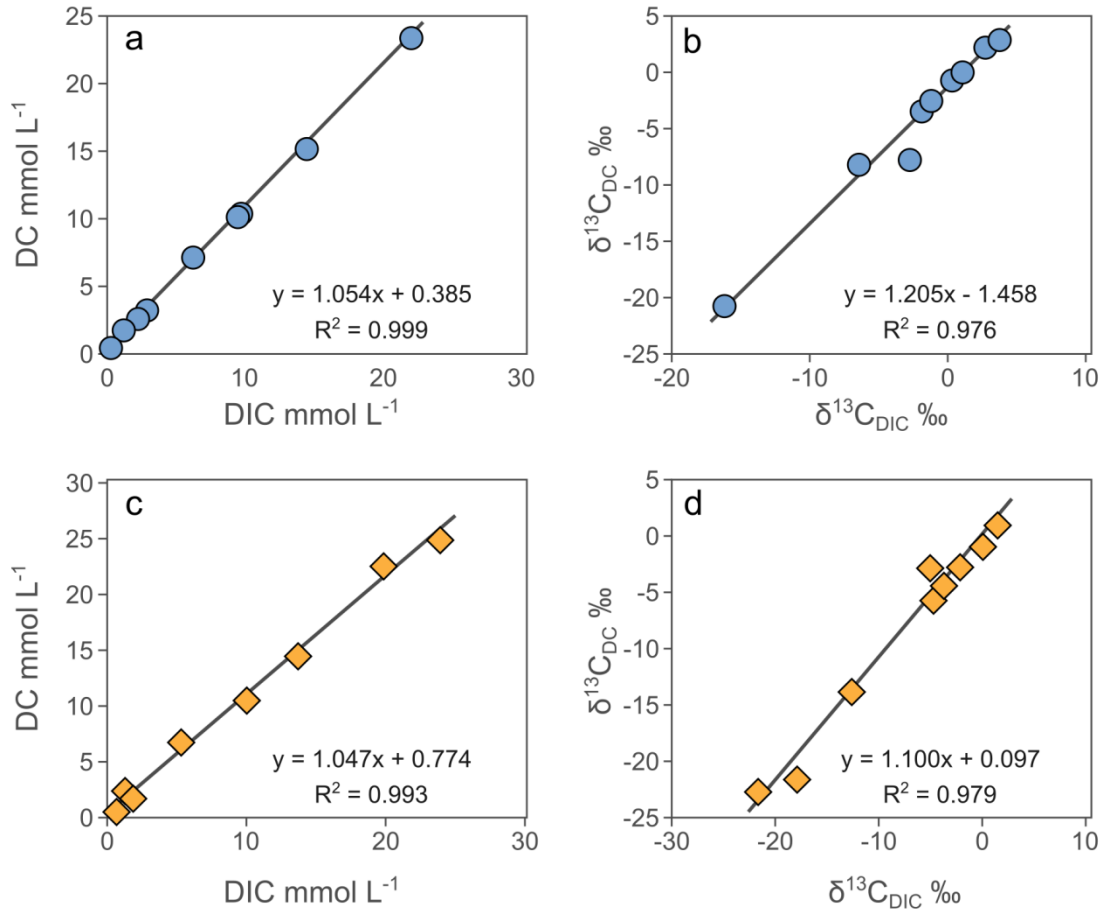
Extended Data Figure 1 – $\text{CO}_2/{}^3\text{He}$ vs. $\delta^{13}\text{C}$ for all samples collected, together with endmember mixing lines between mantle, organic sediment and carbonate endmembers. We argue that such mixing relationships cannot easily explain the water data, and instead carbon isotope fractionation associated with calcite precipitation and chemolithoautotrophy introduces the observed $\delta^{13}\text{C}$ variations.



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1224 **Extended Data Figure 2** – Helium isotopes ($^3\text{He}/^4\text{He}$) vs. X-values (air-normalized
 1225 $^4\text{He}/^{20}\text{Ne}$; considering solubility in water for fluid samples²³). The majority of samples
 1226 have high (>5) X-values, indicating minimal air-contributions to samples.



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1229 **Extended Data Figure 3** – Relationship between DC and DIC concentrations and $\delta^{13}\text{C}$.

1230 Values for northern Costa Rica (panels A and B) are shown with blue symbols and

1231 central Costa Rica (panels c and d), with red symbols. Strong correlations allow

1232 prediction of DC concentrations and $\delta^{13}\text{C}$ values for the sites for which DIC compositions

1233 are lacking. The slope of the concentration plots is used to calculate the fraction of DIC

1234 and DOC in the sample suites.