

1 Variable water input controls evolution of the Lesser Antilles
2 volcanic arc

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4 George F. Cooper^{1*}, Colin G. Macpherson², Jon D. Blundy¹, Benjamin Maunder³, Robert W.
5 Allen³, Saskia Goes³, Jenny Collier³, Lidong Bie⁵, Nick Harmon⁴, Stephen P. Hicks³,
6 Alexander A. Iveson², Julie Prytulak², Andreas Rietbrock^{5,6}, Catherine Rychert⁴, Jon P.
7 Davidson² and the VoiLA team

8
9 ¹ School of Earth Sciences, University of Bristol, Wills Memorial Building, Bristol BS8 1RJ,
10 UK

11 ² Department of Earth Sciences, Durham University, Science Labs, Durham DH1 3LE, UK

12 ³ Department of Earth Science and Engineering, Imperial College London, SW7 2AZ, UK

13 ⁴ University of Southampton, National Oceanography Centre, European Way, Southampton,
14 SO14 3ZH, UK

15 ⁵ Department of Earth Ocean & Ecological Sciences, University of Liverpool, UK

16 ⁶ Geophysical Institute (GPI), Karlsruhe Institute of Technology, 76187 Karlsruhe, Germany

17
18 *corresponding author, email: CooperG3@cardiff.ac.uk, current address: School of Earth and
19 Ocean Sciences, Cardiff University, Cardiff, CF10 3AT, UK

20
21 A list of authors and their affiliations appears at the end of the paper

22
23 **Oceanic lithosphere carries volatiles, notably water, into the mantle via subduction at**
24 **convergent plate boundaries. This subducted water exercises a key control on the**
25 **production of magma, earthquakes, formation of continental crust and mineral**
26 **resources. However, identifying different potential fluid sources (sediments, crust and**

27 **mantle lithosphere) and tracing fluids from their release to observed surface**
28 **expressions has proved challenging¹. The two Atlantic subduction zones are valuable**
29 **end members to study this deep water cycle because hydration in Atlantic lithosphere,**
30 **produced by slow spreading, is expected to be highly non-uniform². As part of an**
31 **integrated, multi-disciplinary project in the Lesser Antilles³, we studied boron trace**
32 **element and isotopic fingerprints of melt inclusions. These reveal that serpentine, i.e.**
33 **hydrated mantle rather than crust or sediments, is a dominant supply of subducted**
34 **water to the central arc. This serpentine is most likely to reside in a set of major**
35 **fracture zones subducted beneath the central arc over the past ~10 Myr. Dehydration**
36 **of these fracture zones is consistent with the locations of the highest rates of**
37 **earthquakes and prominent low shear velocities, as well as time-integrated signals of**
38 **higher volcanic productivity and thicker arc crust. These combined geochemical and**
39 **geophysical data provide the clearest indication to date that the structure and hydration**
40 **of the downgoing plate are directly connected to the evolution of the arc and its**
41 **associated hazards.**

42

43 The 750 km-long Lesser Antilles volcanic arc (LAA), located along the eastern margin of the
44 Caribbean Plate, is the result of slow (1-2 cm/year) westward subduction of Atlantic and
45 proto-Caribbean oceanic lithosphere (Fig 1). Water hosted in hydrous phases within the
46 subducting plate will be released as the slab sinks into the mantle and warms up. As the water
47 migrates out of the slab the stress on faults is reduced, causing earthquakes. At the same time,
48 the addition of water to the overlying mantle wedge reduces the solidus temperature which
49 may enhance melting. LAA magma production rates lie at the lower end of the global range,
50 probably due to the low convergence rates, and are very unevenly distributed, being greatest
51 in the centre of the arc (Dominica and Guadeloupe)⁴. The LAA also displays notable along-

52 arc variations in geochemistry, volcanic activity, crustal structure, and seismicity⁵⁻⁸.
53 Subducting plate velocity and age are often held responsible for variations in convergent
54 margin behaviour⁹ but are unlikely to have first-order influence on lateral variations within
55 the LAA as neither vary significantly along-strike. Instead, variations in LAA magmatism
56 and seismicity have been proposed to reflect; (i) a combination of a strong north to south
57 increase in sediment input¹⁰, (ii) subduction of bathymetric ridges below the central arc¹¹,
58 which may enhance plate stress and coupling, (iii) and/or subduction of strongly hydrated
59 fracture zones¹² at several locations along arc (Fig. 1).

60

61 Current plate reconstructions¹³ show the northern LAA to be underlain by ~90 Ma subducted
62 lithosphere that formed at the Equatorial Mid-Atlantic Ridge and includes the Marathon and
63 Mercurius fracture zones (Fig. 1), whereas beneath the southern LAA, the subducted
64 lithosphere is up to 120 Ma old and formed at the, now-fully subducted, proto-Caribbean
65 mid-ocean ridge. The seafloor spreading rates were slow in both cases. The boundary
66 between the two seafloor-spreading domains is clearly visible in both bathymetric and gravity
67 data, projecting from the Demerera Plateau toward the central islands before becoming
68 obscured by the accretionary prism around Barbados (Fig. 1; Extended Data Fig. 1).

69

70 Hydration of lithosphere formed by intermediate or fast spreading occurs mainly in the mafic
71 crust through faults that form as the plate bends into the trench. By contrast, slow spreading
72 produces highly tectonised oceanic lithosphere with relatively thin mafic crust, pronounced
73 faults, and sections of upper mantle material exposed at the seafloor¹⁴. The transform faults at
74 slow spreading ridges, which manifest as fracture zones in mature oceanic crust, are more
75 seismically active and penetrate to greater depths than in faster-spread lithosphere¹⁵. These
76 large-scale faults provide pathways for seawater and low/medium temperature alteration

77 including hydration of the mantle mineral olivine to serpentine¹⁶. Serpentine, in the form of
78 antigorite, can hold up to 13 wt. % structural water, at least double the water capacity of
79 hydrated mafic crust. Thus, subduction of serpentinized mantle lithosphere has the potential
80 to supply substantial volumes of fluid to magmatic arcs. In order to evaluate along-arc
81 variations of slab-derived fluid sources (e.g. sediment, oceanic crust, or serpentinized mantle
82 lithosphere), we measured trace element concentrations and boron isotopic ratios of melt
83 inclusions along the entire LAA. To investigate how fluids influence arc magma genesis and
84 evolution we compare these geochemical proxies for slab-derived fluids with newly-acquired
85 geophysical data³, and with the predicted positions of subducted fracture zones and the proto-
86 Caribbean/Equatorial Atlantic plate boundary below the arc at different times.

87

88 In subduction zone magmas, boron and its isotopes trace contributions from fluids released
89 by the subducting plate^{17,18}. Boron is fluid mobile, and a high ratio of boron to fluid-
90 immobile elements, like Ti, Nb, or Zr, in arc magmas suggests boron is principally supplied
91 by subducting-plate fluids¹⁹. Serpentine-derived boron is enriched in ¹¹B compared to ¹⁰B,
92 producing distinctively elevated $\delta^{11}\text{B}$ values of +7‰ to +20‰¹⁷ ($\delta^{11}\text{B} =$
93 $((^{11}\text{B}/^{10}\text{B})_{\text{sample}} / (^{11}\text{B}/^{10}\text{B})_{\text{standard}} - 1) \times 10^3$). As a result, arc magmas produced through mantle
94 melting induced by serpentine-derived fluids have significantly higher $\delta^{11}\text{B}$ values (up to
95 +18‰²⁰) than MORB-source mantle ($-7.1 \pm 0.9\%$ ²¹). Fluids derived from subducted
96 sediments have yet a different distinct chemical signature²². Sediments in ocean drill cores
97 east of the LAA contain terrigenous turbidites, pelagic clays, and ashy siliceous clays²³.
98 Although these sediments are enriched in boron (50-160 ppm B), they have significantly
99 lower $\delta^{11}\text{B}$ values (approximately -15 to +5‰²¹) than serpentine-derived fluids at sub-arc
100 depths²⁴.

101

102 Using secondary ion mass spectrometry (SIMS), we measured 198 glassy, clinopyroxene-
103 hosted melt inclusions for volatiles (H₂O, CO₂) and trace elements, of which 92 were further
104 analysed for boron isotopic composition. The analysed melt inclusions are from fresh
105 volcanic deposits assumed to be <<1 million years old (Ma), and range from low-MgO, high-
106 alumina basalt (MgO = 1.8-3.5 wt. %, Al₂O₃ = 15.3-19.1 wt. %) to rhyolite (≤78 wt. % SiO₂;
107 Fig. 2). All of these compositions have undergone some level of magmatic differentiation in
108 the shallow crust, thus none can be considered primary, however, the boron isotopic signature
109 is largely determined by the source rather than subsequent differentiation processes^{25,26}. We
110 supplemented our dataset with all previously published LAA melt inclusion analyses
111 (*n*>1000) available from the GEOROC database.

112
113 LAA melt inclusions are characterised by dissolved water contents of up to 9.1 wt. % H₂O,
114 with a large range for individual islands (Fig. 2). However, water contents of melt inclusions
115 are affected by differentiation processes during crustal storage and thus are a poor proxy for
116 primary magmatic water contents. Water content will increase in a melt undergoing
117 undersaturated crystallisation, remain constant under water saturated conditions, and be lost
118 from melt during late-stage degassing. Further modification of water in melt inclusions can
119 occur due to post entrapment crystallisation and/or diffusive water loss. Ratios of fluid
120 mobile to fluid immobile trace elements, such as B/Nb (Fig. 2), are more reliable indicators
121 of the contribution of fluids, as both elements behave similarly during melting and magmatic
122 differentiation. Our data shows high ratios of B/Nb in the central arc which most probably
123 reflect a particularly fluid and B-rich magmatic source.

124
125 The new δ¹¹B values for LAA melt inclusions vary from -2.8‰ to +11.2‰ (Fig. 2), which
126 spans much of the global arc range (-9‰ to +16‰¹⁷). Melt inclusions with the highest δ¹¹B
127 values are from the central arc (islands of Guadeloupe and Dominica; Fig. 2). δ¹¹B variation

128 within each volcanic centre is unlikely to be due to crustal differentiation because there are
129 no systematic trends in $\delta^{11}\text{B}$ with indicators of differentiation (e.g. SiO_2 and Rb/Sr , Extended
130 Data Fig. 3). This is consistent with prior findings that fractional crystallisation has negligible
131 effect on melt $\delta^{11}\text{B}$ values^{25,26}. Crustal assimilation during open-system differentiation may
132 also modify $\delta^{11}\text{B}$ and B/Nb , but inputs from this source likely have a similar isotopic and
133 geochemical composition to AOC and sediment²². Assimilation of LAA crust would lower
134 melt $\delta^{11}\text{B}$ values during differentiation, a trend that is not observed in our data (Extended
135 Data Fig. 3). Although there is a range of melt inclusion $\delta^{11}\text{B}$ values within each single
136 volcanic centre (e.g. 3.5 ‰ in Martinique) there are clear $\delta^{11}\text{B}$ differences between
137 neighboring volcanic centres with similar major element chemistry. Therefore, we interpret
138 the distinct $\delta^{11}\text{B}$ values in evolved melt inclusions at each island as a reflection of differences
139 between the mantle source regions of each island, such that boron isotopes provide a robust
140 tracer for the fluid source¹⁸.

141

142 We interpret the $\delta^{11}\text{B}$ differences between islands and the systematic $\delta^{11}\text{B}$ change along the
143 arc to result from variable involvement of fluids from two distinct sources: (1) altered
144 oceanic crust (AOC) and sediment; and (2) serpentine dehydration (Fig. 3). In the central
145 portion of the arc, melt inclusions from Guadeloupe and Dominica have $\delta^{11}\text{B}$ values
146 significantly greater than +5‰. Of the available sources, only fluid with > 60% contribution
147 from serpentine dehydration has the capacity to generate this isotopic signature (Fig. 3). The
148 lower $\delta^{11}\text{B}$ values found in the north and south of the arc can be attributed primarily to fluid
149 released by dehydration of AOC and sediment (Fig. 3). However, there is no simple
150 relationship between $\delta^{11}\text{B}$ and indicators of varying volume of fluid addition (e.g. B/Be and
151 B/Nb ; Extended Data Fig. 3). In contrast to Guadeloupe and Dominica, St. Lucia melt
152 inclusions from this study have a high net fluid contribution based on the Nb/B values, but

153 we estimate <30% of this originates from serpentine. Therefore, the total volume of fluid is
154 decoupled from the proportion of different sources from which each fluid is derived. In the
155 north and south of the arc, with the exception of St. Vincent, the proportion of fluid derived
156 from serpentine is lower than in the central arc. Based on boron isotopes it is not possible to
157 distinguish if the serpentinite fluids are derived from the slab or from recycled forearc
158 material^{20,27}. However, a peak in seismicity occurs in the central arc at the depths where
159 models predict dehydration of peridotite in the slab (120-160km)^{9,28}. In conjunction with the
160 abundance of serpentinitised peridotite expected in slow-spread lithosphere^{14,29} this provides
161 an argument for slab-hosted serpentine being the main deliverer of fluid to LAA mantle
162 wedge.

163

164 We compared our geochemical results to a range of independent observations that may be
165 expressions of fluid release (Fig 4). As these observations sample different parts of the
166 subduction system in space and time, we modelled expected excess hydration i.e., fluid
167 derived from fractures zones, to the arc over the past 25 Myr (Fig. 4b), assuming that the
168 known fracture zones and plate boundary between the proto-Caribbean and Atlantic bring
169 extra water in the form of serpentine (see Methods).

170

171 If higher recent fluid fluxes below the arc were to cause an increase in magmas production
172 then we might expect to see boron isotope ratios (Fig 4a) and/or intraslab seismicity rates³⁰
173 correlate with volcanic production rates⁴ (Fig. 4 e and f). Slab seismicity is often attributed to
174 dehydration embrittlement³¹, and the depths to which seismicity extends³⁰ is consistent with
175 the extent of the serpentinite stability field predicted for the convergence rates and ages of
176 LAA subduction. Our data show a peak in boron isotopes, intraslab seismicity rates and
177 volcanic production rates around Dominica, and this is where our forward models (Fig. 4b)

178 predict a peak in dehydration from 0-2 Ma of subduction of the Marathon and Mercurius
179 fracture zones. Therefore, our data indicate that enhanced fluid fluxing of the mantle wedge
180 is associated with higher magma production in the LAA. However, because it is not possible
181 to quantify the relative controls of flux melting versus decompression melting with the
182 available data we cannot identify the cause of any relationship at present.

183

184 High ratios of small to large earthquakes (high b -values) on the plate interface and forearc¹²
185 (Fig. 4c), as well as low shear-wave velocities (4.3 ± 0.05 km/s) at 50 km depth (Fig. 4d,
186 derived from Rayleigh waves recorded during the VoiLA seismic experiment³ - see Methods)
187 could reflect excess dehydration at shallower depths. High b -values are commonly attributed
188 to seismogenic failure at lower stresses due to higher pore fluid pressures, while shear
189 velocity anomalies of around 9% could correspond to about 1.1 vol. % of fluids and
190 associated melts³². Shear velocities and b -values are characterised by a prominent maximum
191 and minimum, respectively, in the region around Martinique, i.e. displaced southward from
192 the peak in boron isotopes. Due to the obliquity of the fracture zones to the trench, excess
193 forearc dehydration (derived from shallower slab depths) is expected to occur further to the
194 south than dehydration below the arc, coincident with the b -value and shear velocity peaks
195 (Fig. 4b).

196

197 Finally, there are systematic variations in crustal thickness along the arc⁷, with thicknesses of
198 around 35 km north of Martinique and around 30 km in the south. These reflect a long-term
199 integrated variation in magma productivity. When we consider the excess dehydration over
200 the age of the present arc (around 25 Myr), the position of Marathon-Mercurius fracture zone
201 subduction has shifted from the north near St Kitts to Dominica today, hence a larger crustal

202 thickness would be expected along the whole northern arc, as observed. Again, however, we
203 cannot constrain the relative role of decompression melting in this magma production.

204

205 None of the other Atlantic fracture zones have contributed to dehydration below the arc. The
206 15-20 fracture zone has not subducted deep enough (but higher b -values and lower shear-
207 wave velocities in the forearc near Antigua in Fig. 4 could, given spatial resolution of these
208 measurements, indicate shallow fluid release from it). Other Atlantic fracture zones have yet
209 to reach the trench. It is likely that there were fracture zones in the Proto-Caribbean oceanic
210 lithosphere but their location is uncertain. We included in our model a single, large-offset
211 fracture zone at the location required to fit the basin geometry between the Bahamas Bank
212 and Demerara Rise (Fig. 1; see Methods). This yields a small peak in excess dehydration in
213 the southernmost arc. Thus, within the uncertainties, Proto-Caribbean fracture zones could
214 explain the increases in $\delta^{11}\text{B}$ and b -values and decrease in shear velocities around St.
215 Vincent and Grenada.

216

217 Given the geological complexity of subduction systems, our new geochemical and
218 geophysical expressions of fluids along the LAA show remarkable coherence with the
219 predicted history of fluid release from fracture zones in the subducting plate at different
220 locations in the system and over different temporal windows. Furthermore, the high boron
221 contents and elevated $\delta^{11}\text{B}$ signature of melt inclusions in magmas from the central segment
222 of the arc are unambiguous indicators of dehydration of subducted serpentine, which is
223 expected to be one the main minerals formed in fracture zone hydration. Therefore, our
224 observations provide strong evidence that a heterogeneous distribution of serpentine in
225 subducting mantle lithosphere exerts a primary control on along-arc variations in mantle

226 wedge hydration and seismicity and may also influence the crustal structure and magmatic
227 productivity of volcanic arcs.

228

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320

321 **End notes**

322 **Data availability statement**

323 All geochemical data generated during this study are included in this published article (and
324 its supplementary information files) and can be accessed in the EarthChem repository
325 (<https://doi.org/XXXX/XXXX>). Compiled geochemical data is freely available from the
326 GEOROC database. Metadata of the VoiLA broadband OBS network and used land stations,
327 a catalogue of the local earthquakes, and teleseismic Rayleigh wave data can be accessed
328 through the Zenodo repository: <https://doi.org/10.5281/zenodo.3725528>. All broadband OBS
329 data collected by the VoiLA project will become freely available through the IRIS DMC
330 (Data Management Center) via their data request tools, at the end of the project (April 2021).

331

332 **VOILA team consortium**

333

334 George F. Cooper¹, Colin G. Macpherson², Jon D. Blundy¹, Benjamin Maunder³, Robert W.
335 Allen³, Saskia Goes³, Jenny Collier³, Lidong Bie⁵, Nick Harmon⁴, Stephen P. Hicks³,
336 Andreas Rietbrock^{5,6}, Catherine Rychert⁴, Jon P. Davidson², Richard G. Davy³, Tim J.
337 Henstock⁴, Michael J. Kendall², David Schlaphorst², Jeroen van Hunen², Jamie J.
338 Wilkinson^{3,7}, Marjorie Wilson⁸

339

340 ⁶Geophysical Institute (GPI), Karlsruhe Institute of Technology, 76187 Karlsruhe, Germany

341 ⁷Department of Earth Sciences, Natural History Museum, Cromwell Road, London SW7

342 5BD, UK

343 ⁸School of Earth and Environment, University of Leeds, Leeds, LS2 9JT, UK

344

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355

356 **Author Contributions**

357 All authors discussed the results and implications of the work and commented on the
358 manuscript at all stages. G.F.C., C.G.M., J.D.B., and A.A.I carried out geochemical analysis
359 and interpretation. G.F.C, S.G., C.G.M, J.D.B., and J.C. drafted the manuscript. N.H. and
360 C.R. produced the shear-wave velocity model. B.M. made the dehydration model. L.B. and
361 S.P.H compiled local seismicity data, D.S. mapped b-values. R.W.A and J.C. produced the
362 tectonic reconstruction and associated figures. C.G.M., S. G., J. D. B., J.C., A.R., N.H., C.R.,
363 J.P.D., T.J.H., J.v.H., J.J.W and M.W designed the original VoiLA experiment.

364

365 **Author Information Statement**

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367 The authors declare no competing interests.

368 Correspondence and requests for materials should be addressed to CooperG3@cardiff.ac.uk

369

370

371 **Figure Captions**

372 **Fig. 1.** Bathymetric map of the study area showing the islands of the Lesser Antilles Arc
373 (LAA, red). Map shows locations of the trench (purple line), oceanic fracture zones (black

374 lines, dashed where subducted), boundary between the proto-Caribbean and equatorial
375 Atlantic seafloor (red line) and South American continent-ocean boundary (yellow line).
376 Proto-Caribbean fracture zones have fully subducted; the likely location of a single one,
377 required by basin geometry, is shown as a light dashed line. The bathymetric contrast
378 between the northern and southern forearc is due to a strong difference in sediment thickness
379 (from a few km in the north to > 15 km in the Barbados accretionary prism). Depth contours
380 of the slab below the LAA are shown every 20 km (light blue lines) and every 100 km (dark
381 blue lines). See Methods and Extended Data Figures 1 and 2 for further details.

382

383 **Fig. 2.** Bathymetric map of the Lesser Antilles Arc compared to water, B/Nb ratios, and $\delta^{11}\text{B}$
384 of melt inclusions in lavas. H_2O (this study and compiled published values) and B/Nb
385 symbols are coloured by the SiO_2 wt% of melt inclusions, as an indicator of magmatic
386 differentiation. $\delta^{11}\text{B}$ symbols are coloured by B/Nb as an indicator of fluid addition.
387 Previously published boron isotope ratios from melt inclusions^{33–35} are shown as crosses.
388 Error bars on $\delta^{11}\text{B}$ values represent propagated 1σ uncertainties and are typically $<\pm 1\%$.

389

390 **Fig. 3.** Melt inclusion Nb/B versus $\delta^{11}\text{B}$ for Lesser Antilles Arc magmas from this study.
391 Mixing model (black lines) shows contamination of depleted mantle (DM, grey square) by
392 fluid derived from serpentinite and from altered oceanic crust (AOC) + sediment-derived
393 fluids at 120 km depth. Green bar represents global serpentinite range. Red and green
394 numbers represent the percentage by mass of fluid from the two sources added to the mantle.
395 Inputs for the model are detailed in Methods. Dotted lines indicate composite fluids formed
396 by mixing between (0.1% and 1% mass) fluids from the two discrete sources. Shading
397 indicates >60% (green), 30-60% (blue), and <30% (yellow) contribution from subducted
398 serpentinite. Darker and lighter shaded areas represent domains referred to in text as ‘high’

399 and 'low' fluid contributions, respectively. Only samples measured in this study are plotted.
400 Error bars on $\delta^{11}\text{B}$ values represent propagated 1σ uncertainties and are smaller than symbol
401 size where absent. All 1σ uncertainties are typically $<\pm 1\%$.

402

403 **Fig. 4.** Summary of along-arc geochemical and geophysical data. (a) Boron isotope ratios of
404 melt inclusions with latitude in the LAA (data symbols coloured as in Fig. 3; previously
405 published data^{33–35} shown by crosses). Light and dark coloured shaded areas correspond to
406 those in Fig. 3. (b) Modelled sub-arc excess (i.e. fracture-zone associated) dehydration
407 averaged over the past 2 Myr (solid red line for fluids released below the arc, dashed yellow
408 line below the forearc) and 25 Myr (dotted blue line, below the arc) (based on plate
409 reconstruction and slab geometry, see Methods). (c) *b*-value distribution (relative frequency
410 of small vs large events below the forearc)¹². (d) Shear-wave velocity from teleseismic
411 Rayleigh waves at 50 km depth, with main anomalies below the forearc. (e) Local seismicity
412 in the subducting plate³⁰. (f) Volcanic production rates over the last 100 kyr as dense-rock-
413 equivalent volumes (DREV)³ (red lines). (g) Crustal thickness below the arc from receiver
414 functions⁷ (blue line). Note how the modelled trends compare well with the main anomalies
415 in data sensitive to recent fluid release below the fore-arc (c,d), below the arc (e,f) and over
416 the past 25 Myr (g).

417

418

419 **Methods**

420

421 **Geochemistry**

422 **a) Sample preparation**

423 Crystals were separated from crushed and sieved scoria, pumice or lava. Picked crystals from
424 the 0.5-1 mm and 1-2 mm size fractions were mounted on glass slides within 2.5 cm diameter
425 aluminium rings, back-filled with epoxy resin, and polished to expose the centre of the
426 crystals. Crystals were imaged under transmitted light to locate the most suitable glassy
427 inclusions before further polishing to expose the maximum number of melt inclusions. All
428 epoxy mounts were gold-coated prior to SIMS analysis.

429

430 **b) Trace elements by SIMS**

431 We measured concentrations of H₂O, CO₂ and trace elements in 198 melt inclusions using the
432 Cameca IMS-4f at the NERC Edinburgh Ion Micro-Probe Facility (EIMF), over two sessions
433 (October 2017 and January 2018). The IMS-4f instrument was run with a 15 kV (nominal)
434 primary beam of O⁻ ions with a beam current of ~5 nA, resulting in a spot size at the sample
435 surface of ~15 µm diameter. Positive secondary ions were extracted at 4.5 kV, using energy
436 filtering with an energy window of 50±25 eV (for CO₂ analysis) or 75±25 eV (for all other
437 elements). CO₂ measurements were performed first. Prior to each analysis, the sample was
438 pre-sputtered using a primary beam raster of 20 µm for 4 minutes to reduce C backgrounds
439 resulting from surface contamination. The isotopes ¹²Mg²⁺, ¹²C, ²⁶Mg, and ³⁰Si were
440 measured. Peak positions were verified at the start of each analysis. The background C signal
441 was determined through analysis of the nominally C-free KL2-G glass standard. Following
442 CO₂ analysis, H₂O and trace element concentrations were measured on the same analytical
443 spot as the CO₂ analyses, using a secondary accelerating voltage of 4500 V with 75 V offset
444 and a 25 µm image field. The isotopes ¹H, ⁷Li, ¹¹B, ¹⁹F, ²⁶Mg, ³⁵Cl, ³⁰Si, ⁴²Ca, ⁴⁴Ca, ⁴⁵Sc,
445 ⁴⁷Ti, ⁸⁴Sr, ⁸⁵Rb, ⁸⁸Sr, ⁸⁹Y, ⁹⁰Zr, ⁹³Nb, ¹³³Cs, ¹³⁸Ba, ¹³⁹La, ¹⁴⁰Ce, and ¹⁴⁹Sm were measured.
446 Calibration was carried out on a range of basaltic glass standards with 0–4 wt.% H₂O,
447 repeated throughout the day. Absolute element concentrations were calculated using the in-

448 house JCION5 software and by normalizing the intensities to Si (as measured using ^{30}Si)
449 which was determined by subsequent electron microprobe analysis. A summary of repeat
450 analyses of GSD-1G and T1-G are presented in the Supplementary Data.

451

452 **c) Electron microprobe**

453 Following volatile and trace element analysis, we measured major elements using a Cameca
454 SX100 electron microprobe (EPMA) at the University of Bristol, UK. The gold coat was
455 removed and samples were carbon-coated. Concentrations of SiO_2 , TiO_2 , Al_2O_3 , Fe_2O_3 ,
456 MnO , MgO , CaO , Na_2O , K_2O , P_2O_5 , Cr_2O_3 , SO_2 , and Cl in glass were made with a 20 kV
457 accelerating voltage, a 4 nA beam current and a 5 μm or 10 μm defocused beam to minimise
458 alkali loss³⁶. Major elements were calibrated using a range of synthetic oxide, mineral and
459 metal standards.

460

461 **d) Boron isotopes by SIMS**

462 Prior to boron isotope analysis, crystals hosting the measured melt inclusions were cut out of
463 the epoxy mounts and pressed into indium within 24 mm diameter Al holders. This step
464 reduced the total number of sample mounts and, as indium outgasses less than epoxy, reduces
465 the time required to reach a suitable vacuum for analysis.

466

467 We measured boron isotopes (^{11}B and ^{10}B) in 92 melt inclusions using the Cameca IMS-1270
468 at the NERC Edinburgh Ion Micro-Probe Facility (EIMF), in December 2018. Prior to
469 analysis, the samples were cleaned and a gold coat was applied. Positive secondary ions of
470 $^{10}\text{B}^+$ and $^{11}\text{B}^+$ were produced by sputtering the sample with a 5nA, $^{16}\text{O}^{2-}$ primary beam with a
471 net impact energy of 22 keV, focused using Köhler illumination to a $\sim 25\mu\text{m}$ spot size.

472 Secondary ions were extracted at 10 kV and counted by a single electron multiplier detector.

473 No energy filtering was applied. Analyses were performed using a mass resolution ($M/\Delta M$)
474 of ~ 2400 . Single analyses consisted of 50 measurement cycles of ^{10}B and ^{11}B signals, using
475 counting times of 2 s. Instrumental fractionation was determined using the reference
476 materials GSD1-G, B6, GOR132-G, StHs6/80-G and BCR2-G, measured at the beginning,
477 during and end of the session (Supplementary Data).

478

479 **Boron mixing model**

480 Element contents for AOC and sediment and serpentinite-derived fluids are from ref.²⁰.
481 Isotope ratios used for serpentinite fluids lie within the range of Atlantic peridotites^{29,37–39}
482 . Depleted Mantle boron concentrations and isotope ratios are from ref.⁴⁰; Nb concentrations
483 are from ref.⁴¹. Values are presented in Extended Data Table 1. Composite fluids are
484 produced by mixing the two most significant endmembers in the Lesser Antilles (AOC +
485 sediment and serpentinite derived fluid).

486

487 **Shear velocity**

488 The ocean-bottom seismic data analysed in this study were collected during two cruises
489 aboard the RRS James Cook^{42,43}. We used vertical seismograms to measure the amplitude
490 and phase of ambient noise cross correlation function and teleseismic Rayleigh Waves. The
491 onshore and offshore data were corrected for instrument response, detrended and demeaned
492 prior to processing. The teleseismic data were further processed as detailed in ref.⁹.
493 Measurements of Rayleigh wave dispersion and estimates of the amplitude at selected period
494 were made using frequency-time analysis^{44,45}. We measured dispersion from 18-11 s period.
495 We used up to 2486 dispersion measurements from 93 events from teleseismic Rayleigh
496 waves in the tomography.

497

498 Shear velocity tomography was performed in two steps: first the amplitude and phase data
499 were inverted for phase velocity maps⁴⁶⁻⁴⁸ and then at each location in the phase velocity
500 maps we inverted for 1D shear velocity structure to generate a 3-D volume⁴⁶. For the shear
501 velocity inversion, we included the effects of the water column and sediment using *a priori*
502 information; our initial crustal thickness was based on Airy isostasy across the region. The
503 shear velocity inversion subsequently solved for the best fitting crustal thickness as well as
504 shear velocity.

505

506 **Plate reconstruction and hydration modelling**

507 **a) Mapping the tectonic features**

508 Our modelling of the subducted features below the Lesser Antilles is based upon the global
509 plate reconstruction of ref.¹³ as implemented within the software G-Plates 2.1. In this
510 reconstruction, the opening of the proto-Caribbean seaway occurs from 150 Ma through
511 symmetrical seafloor spreading between the diverging North American and South
512 America/African plates. For ease of reference, we will refer to this stage as the “proto-
513 Caribbean and central Atlantic” opening. Breakup between the South American and African
514 plates starts around 100 Ma with northward propagation from the south Atlantic. We refer to
515 this second stage of seafloor spreading as “equatorial Atlantic” opening.

516

517 Most of the proto-Caribbean oceanic lithosphere has been subducted, but there remains a
518 small segment in the south of the study area. The rifted oceanic lithosphere boundary
519 between it and the equatorial Atlantic is visible in satellite gravity to the north-west of the
520 Demerara Rise where it clearly acts as the termination point for a number of small fracture
521 zones south of Doldrums Fracture Zone (red ellipse, Extended Data Fig 1b).

522

523 We first compared major Atlantic fracture zones in the region (15-20, Marathon, Mercurius,
524 Vema and Doldrums) as detected in satellite gravity data to modelled flow lines according to
525 the Müller et al. (2019) model (Extended Data Fig 1). Overall, the largest misfit between the
526 two was ~50 km, and we assign this value to the positional uncertainty of these features (see
527 below). The geometrical relationships between the two phases of seafloor spreading are
528 particularly clear on the African side of the Atlantic, where the sediment cover is thin and the
529 full sequence preserved (compared to the sedimented and partially subducted American side).
530 The analysis showed that the southern two fracture zones (Vema and Doldrums) have only
531 just reached the Lesser Antilles trench, whereas the northern fracture zone (15-20) only
532 grazes the Lesser Antilles subduction zone. None of these three fracture zones are therefore
533 sources of hydration below the Lesser Antilles Arc.

534

535 Next we refined the location of the proto-Caribbean / equatorial Atlantic Ocean boundary
536 through time (Extended Data Fig.2) based upon two observations. 1) The oldest section of the
537 Marathon and Mercurius fracture zones can be well fitted by a flowline based entirely upon
538 relative motion between North America and Africa. Therefore, this region must have lain
539 entirely north of (or upon) the boundary between the central Atlantic and proto-Caribbean
540 prior to opening of the equatorial Atlantic. 2) The major fracture zones to the south (Vema
541 and Doldrums) can be well fitted by a flowline based entirely upon relative motion between
542 South America and Africa. In this case, the far western extent of these fracture zones (which
543 is constrained by symmetry with the clearly observable extent of fracture zones on the
544 African side) must mark the edge of the proto-Caribbean oceanic crust in order for the
545 Demerara Rise to close back against the African continental margin prior to initiation of
546 equatorial Atlantic spreading (Extended Data Fig 2a). Finally, the proto-Caribbean spreading

547 ridge was placed mid-way between the separating North and South America plates, with a
548 minimum number of transform faults inserted to satisfy the continental plate geometries.
549 Using this updated geometry for the proto-Caribbean / equatorial Atlantic boundary, and our
550 computed flowlines for the Marathon, Mercurius and unnamed proto-Caribbean fracture
551 zones, we model the subduction of these incoming plate features beneath the Caribbean plate
552 from 50 Ma through to the present day. Convergence azimuths and velocities between the
553 Caribbean plate and the Atlantic are extracted directly from the model of ref.¹³.

554

555 **b) Projecting tectonic features onto the slab**

556 To properly track the features once they enter the subduction zone and the slab begins to dip,
557 it is necessary to adjust their horizontal velocities. To do this, we use three different
558 assumptions for how the slab deforms as it enters the subduction zone. One end-member is
559 the “kinematic” approach outlined in ref.⁴⁹ whereby features are assumed to follow
560 streamlines over the surface of a slab with a fixed geometry, i.e. minimal to no plate
561 stretching during subduction. We use the slab geometry of ref.³⁰ determined using local
562 seismicity, and ref.⁵⁰, which is based on teleseismic tomography, for the regions that this first
563 model does not cover. We also assume that the slab geometry remains fixed relative to the
564 Caribbean plate for the modelled time period. In the other end-member, the slab is assumed
565 to maintain its horizontal velocity and acquire an additional vertical sinking velocity, which
566 would imply some amount of plate stretching. For the plate motions of the region, the first
567 approach places incoming plate features further south than the second. We run a third, “best-
568 estimate” model that is intermediate between the two.

569

570 **c) Dehydration modelling**

571 As incoming plate features move into the subduction zone, they dehydrate. Major pulses of
572 subducting-plate dehydration occur⁹ below the forearc and at subarc depths. Forearc
573 dehydration includes the expulsion of pore fluids and the first breakdown of hydrous phases
574 in the oceanic crust, while the subarc pulse starts with the blueshist transition that initiates
575 directly below the maximum decoupling depth, below which the cool subducting plate first
576 becomes coupled to the hot convecting mantle wedge. Following ref.¹ in computing phase
577 stability fields, and using the kinematic thermal model set up of ref.⁵¹ to compute a thermal
578 structure for the geometry and velocity of the Antilles slab, we predict that the first pulse of
579 dehydration extends down to about 40 km depth, and the subarc pulse peaks at a depth up to
580 100-120 km (based on preliminary tomographic models by ref.⁵²). In a similar model for the
581 Greek subduction zone (which is similarly slow and old as the Antilles), the main
582 dehydration depth intervals agree with regions of high Vp/Vs above the slab, as expected
583 from fluid release⁵³. Motivated by these thermal models, sub-arc observations (number of
584 Benioff zone earthquakes) and observations at the volcanic arc itself (boron isotopic
585 signature, present day volcanic output and crustal thickness) are compared at a dehydration
586 depth of 100 km, which matches the average sub-arc slab depth. Comparisons with
587 observations that reflect conditions beneath the fore-arc (forearc Vs and b-value anomalies),
588 are done at a dehydration depth of 40 km.

589

590 For this study, our interest is in lateral variations in water input. We assume that the fracture
591 zones and Atlantic-Proto-Caribbean boundary are all sources of excess slab hydration, i.e.
592 where the slab incorporates significantly larger quantities of water, mainly in the form of
593 serpentinite, than in the plate away from the fracture zones., based on observations of similar
594 structures offshore central America⁵⁴. In the modelling, we apply the same Gaussian excess
595 hydration profile with a width of 15 km to all these features (i.e. in addition to the uniform

596 background). This width is informed by the lateral extent of the Vp/Vs anomaly observed
597 underneath the Marathon fracture zone on the incoming plate⁵⁶. To put a very approximate,
598 order-of-magnitude estimate on the absolute values for the rate of excess hydration along the
599 arc due to the subduction of each feature, we assume that the region of anomalous Vp/Vs
600 corresponds to 50% serpentinised mantle lithosphere, and that half of this additional water is
601 released under the fore-arc and half under the arc. We only model the along strike-variations
602 in excess dehydration (i.e. we set background hydration to zero).

603

604 We ultimately use the models to calculate the relative rate of hydration along the arc over the
605 past 2 Myr for meaningful comparison with features that should depend on the present
606 day/recent dehydration below the arc and fore-arc, and over the past 25 Myr (the age of the
607 current arc) for meaningful comparison with features that should depend on the total amount
608 of water supplied to the arc (i.e. the crustal thickness). The results of these calculations are
609 presented in Extended Data Fig. 4 for a “best estimate” calculation which uses the “halfway”
610 approach to slab deformation; a “southern bound” calculation, which uses the stretched-slab
611 end member plus a 50 km shift to the south (the maximum misfit between our modelled
612 fracture zones and the actual fracture zones on the African side of the Atlantic); and a
613 “northern bound” model which uses the “minimal-stretching” approach⁴⁹ plus a 50 km shift
614 to the north.

615

616 **d) Key results**

617 If we take the best estimate model, we predict that the dehydration peak due to the Marathon
618 and Mercurius fracture zones and the Proto-Caribbean / equatorial Atlantic plate boundary
619 lies currently underneath Dominica (solid red line). In the main article, we demonstrate that
620 this corresponds well with the peak in $\delta^{11}\text{B}$, sub-arc Wadati-Benioff earthquakes and volcanic

621 output. We also predict that, if these three features are dehydrating underneath the fore-arc,
622 then they would currently be doing so trenchwards of Martinique (dashed yellow line). This
623 corresponds well with anomalies in Vs at a depth of around 50 km and the *b*-values for
624 earthquakes in the fore-arc/plate-interface region. Looking at the full history of the arc (0-25
625 Ma: dotted blue line), there is a broad peak between Dominica and St. Kitts and Nevis; the
626 northern part of the arc. This higher rate of fluid flux in the north of the arc throughout the
627 lifetime of the current arc may have resulted in a higher long-term magmatic output and
628 therefore, a thicker crust⁷ if flux melting occurred. However, we cannot constrain the relative
629 contribution of flux melting versus decompression melting. There are also peaks in the
630 present-day dehydration rate and long-term dehydration rate in the far south of the arc
631 between Grenada and St. Vincent. These are due to the subduction of the unnamed proto-
632 Caribbean fracture zone, the exact position of which is more speculative than for the Atlantic
633 features. However, such features on the proto-Caribbean plate could potentially be
634 responsible for the $\delta^{11}\text{B}$ anomaly observed at St. Vincent.

635

636

637 **References for methods**

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- 691
- 692
- 693
- 694

695 **Extended Data**

696

697 **Extended Data Fig. 1.** a) Modelled fracture zones in the central Atlantic, overlain on an
698 oceanic crust age grid from ref.¹³. Coloured stars denote conjugate points associated with
699 opening of the equatorial Atlantic at either end of the Vema (green) and Doldrums (yellow)
700 fracture zones and between the Demerara rise and African continental margin (red). b)
701 Modelled fracture zones overlain on satellite free-air gravity⁵⁵. Red ellipse marks the location
702 of the proto-Caribbean / Atlantic boundary.

703

704 **Extended Data Fig. 2.** Snap shot of modified plate reconstruction at 50Ma¹³. Velocity
705 vectors (coloured by plate) shown are relative to the mantle reference frame. The figure
706 shows the four sources of dehydration from the subducted slab over the past 25 Ma
707 considered here ((i) Marathon FZ; (ii) Mercurius FZ; (iii) proto-Caribbean/ equatorial
708 Atlantic boundary and (iv) unnamed FZ formed during proto-Caribbean opening – labelled
709 PCFracture Zone)

710

711 **Extended Data Fig. 3.** All melt inclusion $\delta^{11}\text{B}$ values measured in this study versus
712 indicators of fluid composition (a, b), and differentiation (c-e). No clear observable trends are
713 shown between islands, indicating that these differences are largely controlled by the mantle
714 source.

715

716 **Extended Data Fig. 4.** The average rate of excess-dehydration (above a uniform
717 background), resulting from the subduction of fracture zones and the proto-Caribbean /
718 Atlantic plate boundary, along the arc from 11° N to 18° N over the past 2 Myr (red solid
719 curve) and 25 Myr (blue dotted curve), and below the fore-arc over the past 2 Myr (dashed

720 yellow line). The pattern of relative distribution of dehydration is robust, constrained by the
721 history of fracture-zone/plate-boundary subduction, but the absolute values of the
722 dehydration rates should be treated with caution, as they depend strongly on the simple model
723 assumptions of the level of hydration and relative strength of fore- and sub-arc dehydration.
724 Panel (a) is the best estimate (b) is the “northern bound” end-member and (c) is the “southern
725 bound” (see text for details).

726

727 **Extended Data Table 1.** $\delta^{11}\text{B}$ values, B concentrations, and Nb/B of sources of fluids used in
728 the mixing model (Fig. 3).







