

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

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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 challenging<sup>1</sup>. 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-uniform<sup>2</sup>. As part of an  
31 integrated, multi-disciplinary project in the Lesser Antilles<sup>3</sup>, 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)<sup>4</sup>. The LAA also displays notable along-

52 arc variations in geochemistry, volcanic activity, crustal structure, and seismicity<sup>5-8</sup>.  
53 Subducting plate velocity and age are often held responsible for variations in convergent  
54 margin behaviour<sup>9</sup> 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 input<sup>10</sup>, (ii) subduction of bathymetric ridges below the central arc<sup>11</sup>,  
58 which may enhance plate stress and coupling, (iii) and/or subduction of strongly hydrated  
59 fracture zones<sup>12</sup> at several locations along arc (Fig. 1).

60

61 Current plate reconstructions<sup>13</sup> 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 seafloor<sup>14</sup>. 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 lithosphere<sup>15</sup>. These  
76 large-scale faults provide pathways for seawater and low/medium temperature alteration

77 including hydration of the mantle mineral olivine to serpentine<sup>16</sup>. 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 data<sup>3</sup>, 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<sup>17,18</sup>. 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 fluids<sup>19</sup>. Serpentine-derived boron is enriched in <sup>11</sup>B compared to <sup>10</sup>B,  
92 producing distinctively elevated  $\delta^{11}\text{B}$  values of +7‰ to +20‰<sup>17</sup> ( $\delta^{11}\text{B} =$   
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‰<sup>20</sup>) than MORB-source mantle ( $-7.1 \pm 0.9\%$ <sup>21</sup>). Fluids derived from subducted  
96 sediments have yet a different distinct chemical signature<sup>22</sup>. Sediments in ocean drill cores  
97 east of the LAA contain terrigenous turbidites, pelagic clays, and ashy siliceous clays<sup>23</sup>.  
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‰<sup>21</sup>) than serpentine-derived fluids at sub-arc  
100 depths<sup>24</sup>.

101

102 Using secondary ion mass spectrometry (SIMS), we measured 198 glassy, clinopyroxene-  
103 hosted melt inclusions for volatiles (H<sub>2</sub>O, CO<sub>2</sub>) 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. %, Al<sub>2</sub>O<sub>3</sub> = 15.3-19.1 wt. %) to rhyolite (≤78 wt. % SiO<sub>2</sub>;  
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<sup>25,26</sup>. 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. % H<sub>2</sub>O,  
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 δ<sup>11</sup>B 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‰<sup>17</sup>). Melt inclusions with the highest δ<sup>11</sup>B  
127 values are from the central arc (islands of Guadeloupe and Dominica; Fig. 2). δ<sup>11</sup>B 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.  $\text{SiO}_2$  and  $\text{Rb/Sr}$ , 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<sup>25,26</sup>. Crustal assimilation during open-system differentiation may  
132 also modify  $\delta^{11}\text{B}$  and  $\text{B/Nb}$ , but inputs from this source likely have a similar isotopic and  
133 geochemical composition to AOC and sediment<sup>22</sup>. 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 source<sup>18</sup>.

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.  $\text{B/Be}$  and  
151  $\text{B/Nb}$ ; 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  $\text{Nb/B}$  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<sup>20,27</sup>. 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)<sup>9,28</sup>. In conjunction with the  
160 abundance of serpentinitised peridotite expected in slow-spread lithosphere<sup>14,29</sup> 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 rates<sup>30</sup>  
173 correlate with volcanic production rates<sup>4</sup> (Fig. 4 e and f). Slab seismicity is often attributed to  
174 dehydration embrittlement<sup>31</sup>, and the depths to which seismicity extends<sup>30</sup> 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 forearc<sup>12</sup>  
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 experiment<sup>3</sup> - 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 melts<sup>32</sup>. 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 arc<sup>7</sup>, 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**

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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.

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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.  $\text{H}_2\text{O}$  (this study and compiled published values) and B/Nb  
385 symbols are coloured by the  $\text{SiO}_2$  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<sup>33–35</sup> are shown as crosses.  
388 Error bars on  $\delta^{11}\text{B}$  values represent propagated  $1\sigma$  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  $1\sigma$  uncertainties and are smaller than symbol  
401 size where absent. All  $1\sigma$  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<sup>33–35</sup> 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)<sup>12</sup>. (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 plate<sup>30</sup>. (f) Volcanic production rates over the last 100 kyr as dense-rock-  
413 equivalent volumes (DREV)<sup>3</sup> (red lines). (g) Crustal thickness below the arc from receiver  
414 functions<sup>7</sup> (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 H<sub>2</sub>O, CO<sub>2</sub> 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<sup>-</sup> 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 CO<sub>2</sub> analysis) or 75±25 eV (for all other  
437 elements). CO<sub>2</sub> 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 <sup>12</sup>Mg<sup>2+</sup>, <sup>12</sup>C, <sup>26</sup>Mg, and <sup>30</sup>Si 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 CO<sub>2</sub> analysis, H<sub>2</sub>O and trace element concentrations were measured on the same analytical  
443 spot as the CO<sub>2</sub> analyses, using a secondary accelerating voltage of 4500 V with 75 V offset  
444 and a 25 µm image field. The isotopes <sup>1</sup>H, <sup>7</sup>Li, <sup>11</sup>B, <sup>19</sup>F, <sup>26</sup>Mg, <sup>35</sup>Cl, <sup>30</sup>Si, <sup>42</sup>Ca, <sup>44</sup>Ca, <sup>45</sup>Sc,  
445 <sup>47</sup>Ti, <sup>84</sup>Sr, <sup>85</sup>Rb, <sup>88</sup>Sr, <sup>89</sup>Y, <sup>90</sup>Zr, <sup>93</sup>Nb, <sup>133</sup>Cs, <sup>138</sup>Ba, <sup>139</sup>La, <sup>140</sup>Ce, and <sup>149</sup>Sm were measured.  
446 Calibration was carried out on a range of basaltic glass standards with 0–4 wt.% H<sub>2</sub>O,  
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  $^{30}\text{Si}$ )  
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  $\text{SiO}_2$ ,  $\text{TiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{Fe}_2\text{O}_3$ ,  
456  $\text{MnO}$ ,  $\text{MgO}$ ,  $\text{CaO}$ ,  $\text{Na}_2\text{O}$ ,  $\text{K}_2\text{O}$ ,  $\text{P}_2\text{O}_5$ ,  $\text{Cr}_2\text{O}_3$ ,  $\text{SO}_2$ , and Cl in glass were made with a 20 kV  
457 accelerating voltage, a 4 nA beam current and a 5  $\mu\text{m}$  or 10  $\mu\text{m}$  defocused beam to minimise  
458 alkali loss<sup>36</sup>. 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 ( $^{11}\text{B}$  and  $^{10}\text{B}$ ) 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  $\sim 2400$ . Single analyses consisted of 50 measurement cycles of  $^{10}\text{B}$  and  $^{11}\text{B}$  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.<sup>20</sup>.  
481 Isotope ratios used for serpentinite fluids lie within the range of Atlantic peridotites<sup>29,37–39</sup>  
482 . Depleted Mantle boron concentrations and isotope ratios are from ref.<sup>40</sup>; Nb concentrations  
483 are from ref.<sup>41</sup>. 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<sup>42,43</sup>. 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.<sup>9</sup>.  
493 Measurements of Rayleigh wave dispersion and estimates of the amplitude at selected period  
494 were made using frequency-time analysis<sup>44,45</sup>. 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 maps<sup>46-48</sup> and then at each location in the phase velocity  
500 maps we inverted for 1D shear velocity structure to generate a 3-D volume<sup>46</sup>. 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.<sup>13</sup> 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.<sup>13</sup>.

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.<sup>49</sup> 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.<sup>30</sup> determined using local  
562 seismicity, and ref.<sup>50</sup>, 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 occur<sup>9</sup> 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.<sup>1</sup> in computing phase  
577 stability fields, and using the kinematic thermal model set up of ref.<sup>51</sup> 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.<sup>52</sup>). 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 release<sup>53</sup>. 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 America<sup>54</sup>. 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 plate<sup>56</sup>. 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” approach<sup>49</sup> 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  $V_s$  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 crust<sup>7</sup> 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

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695 **Extended Data**

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697 **Extended Data Fig. 1.** a) Modelled fracture zones in the central Atlantic, overlain on an  
698 oceanic crust age grid from ref.<sup>13</sup>. 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 gravity<sup>55</sup>. 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 50Ma<sup>13</sup>. 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).









