

Carbonate crusts have been recovered on the slopes of volcanic islands using remotely operated vehicles

Crusts consist of mixtures of volcanoclastic and biogenic fragments cemented together by calcite in various forms

Crusts are attributed to seawater circulating within the surficial sediment, in some cases mixing with hydrothermal fluids based on isotopic analyses

Crusts provide increased stability of submarine slopes by armorizing surficial sediments

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Carbonate crusts around volcanic islands and their significance

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37 ABSTRACT

38 Extensive carbonate crusts discovered forming on the slopes of seamounts in many parts of
39 the world's oceans are providing extra stability to the volcanic edifices. These crusts are
40 composed of mixtures of volcanoclastic debris and bioclastic material, in most cases
41 cemented by calcite, in the form of isopachous coatings around grains and pore-filling spar.
42 Such crusts, which have been collected by remotely-operated vehicle (ROV), are described
43 here from moderate-depth to deeper-water slopes (200 – 1000 m) around the volcanic island
44 of Montserrat in the Caribbean, and from the nearby Kick'em Jenny submarine volcano off
45 Grenada. Radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratios from the carbonates give an indication of age but
46 also demonstrate that some samples have been altered by hydrothermal-volcanic processes, as
47 shown by ages much older than expected. Such alteration is also supported by carbon and
48 oxygen isotope ($\delta^{13}\text{C}$ and $\delta^{18}\text{O}$) ratios, although most samples have typical marine values. In
49 many cases $\delta^{18}\text{O}$ is usually a little more positive than expected from modern shallow-water
50 carbonates, likely reflecting cooler water at the depth of lithification. Just one sample, from
51 Kick'em Jenny, has very negative $\delta^{13}\text{C}$ (-42 ‰) indicating methanogenesis. Crusts are also
52 reported here from the Mediterranean Sea, with an example described from Kolumbo,
53 northeast of the Santorini volcanic complex in the Hellenic subduction zone, that are similar
54 in many respects to those from the Caribbean. Typically, the biota of the crusts consists of
55 calcareous red algae (commonly encrusting volcanic clasts), benthic (also encrusting) and
56 planktic foraminifera, subordinate serpulids, bivalves, pteropods and heteropods, and rare
57 deeper-water corals. In addition, there is evidence for the former presence of microbes from
58 the occurrence of calcified filaments and peloids in intragranular cavities. Several generations
59 of sponge borings are usually present as well as calcite cement. The carbonate crusts are
60 attributed to seawater circulating within the surficial sediment, in some cases mixing with
61 hydrothermal fluid. The significance of these crusts is in stabilising seamounts, enabling their

62 slopes to avoid frequent collapse, dissection and readjustment, but when failure does occur,
63 larger-scale submarine landslides involving coherent slabs are more likely.

64

65 Keywords: Seamounts, Montserrat, hardgrounds, carbonate crusts, slope crusts,

66

67 **1. Introduction**

68 Volcanic islands are favourable environments for the formation of carbonate sediment.

69 Much attention has focussed on the formation of coral islands and atolls by the subsidence of
70 volcanic islands following on from Darwin's seminal work (Darwin, 1874; Stoddart, 1976).

71 Coral reefs surround volcanic islands and protect them from erosion by waves and storms.

72 They are generated by biological activity but cementation is also involved due to interaction
73 with carbonate-saturated fluids, notably seawater. We have discovered extensive carbonate-

74 cemented crusts that mantle and armour the deeper flanks of volcanic islands and seamounts
75 as submarine hardgrounds to water depths of several hundred metres. The hardgrounds form

76 by cementation of volcanoclastic and bioclastic deposits that drape seamount slopes. Although
77 hardgrounds are relatively common in shallow-water successions throughout the geological

78 record, and relatively rare in deeper-water facies (Christie et al., 2015), the term

79 "hardground" was first introduced by Murray and Renard (1891) as a descriptive term for

80 rocky seafloors in the deep-sea discovered during the voyage of HMS Challenger in 1872-6.

81 'Classic' modern-Quaternary shallow-water hardgrounds are well-documented from

82 carbonate-dominated locations such as the Bahamas and Arabian Gulf (e.g. Shinn, 1968;

83 Dravis, 1978) and deeper-water ones occur there on slopes too (e.g. Schlager and James,

84 1978; Malone et al., 2001), as well as in the Mediterranean (Allouc, 1990; Aghib et al., 1991)

85 and on cold-water mound structures, such as Porcupine Bank, in the NE Atlantic (Noé et al.,

86 2006; van der Land et al., 2010). There are, however, fewer reports of carbonate crusts on the

87 slopes of seamounts and volcanic ridges, apart from some documented from the Mid-Atlantic
88 Ridge (Thompson et al., 1968; Schroeder et al., 2002), the Mediterranean (McKenzie and
89 Bernoulli, 1982) and Red Sea (e.g., Milliman, 1974). Several Quaternary hardgrounds have
90 been related to sea-level and climate changes (see reviews of van der Kooij et al., 2010 and
91 Christ et al., 2015).

92 Exploration of the submarine flanks of active volcanoes and volcanic seamounts during
93 expeditions of the EV *Nautilus* has revealed common limestone crusts and hardgrounds
94 covering their submarine slopes. These crusts typically consist of volcanogenic clastic and
95 biogenic material cemented by calcite coating the slopes and extending to water depths of
96 several hundred metres. They can be several tens of centimetres to a few metres thick and
97 form distinctive slabs when fractured. They have been observed at the Soufrière Hills
98 Volcano, Montserrat in the Eastern Caribbean, and around several seamounts and young
99 submarine volcanoes in the Mediterranean and in the Galapagos Islands, all of which have
100 been sampled. To our knowledge such carbonate crusts have not been widely recognised or
101 described. However, they may be significant from several perspectives. First, they represent a
102 new location of limestone deposition in the world's oceans. Second, the hardgrounds act to
103 protect the submarine flanks of volcanic islands and seamounts from erosion and collapse and
104 they may inhibit submarine intrusion of seawater into the interior of such volcanic edifices. In
105 addition, they also form important hard substrates for the development of benthic biological
106 communities.

107 This study focuses on the development of limestone crusts with a case study of the Soufrière
108 Hills Volcano (SHV), Montserrat, together with descriptions of occurrences on other
109 seamounts, in the southern Lesser Antilles (Kick'em Jenny submarine volcano) and, as a
110 Mediterranean example, the Kolumbo seamounts from the Hellenic Arc in the Aegean Sea.
111 We also note limestones sampled on Montserrat in geothermal boreholes and as clasts in

112 debris-avalanche deposits since they are pertinent to understanding the role of hydrothermal
113 and ground-water systems in forming the limestone crusts. The origin of the seamount-slope
114 limestones is elucidated through petrography, geochronology and isotope geochemical
115 studies. Important questions to be addressed include: what are the relative roles of inorganic
116 versus organic processes in the formation of the crusts; what are their rates of formation, and
117 what environmental factors such as water depth, temperature and fluid-flow related to
118 hydrothermal systems, are important controlling factors in their occurrence.

119

120 **2. Geological Settings**

121 *2.1 The Soufrière Hills Volcano, Montserrat*

122 Montserrat is a predominantly andesitic volcanic island in the eastern Caribbean that is
123 elevated to a height of almost 2 km above the surrounding seafloor from water depths of 800
124 to 1000 m. The island comprises four volcanic centres (Fig. 1) which become younger from
125 north to south (Rea, 1974; Harford et al., 2002; Coussens et al., 2017; Hatter et al., 2018).

126 These centres are the Silver Hills (2.17 to 1.03 Ma), Centre Hills (1.14 to 0.38 Ma), South
127 Soufrière Hills (0.14 to 0.09 Ma) and the Soufrière Hills Volcano (SHV, 0.45 Ma to present).

128 The major eruption of the SHV between 1995-2010 (Kokelaar, 2002; Wadge et al., 2014) has
129 resulted in Montserrat being one of the most intensively studied island-arc volcanoes in the
130 World. A striking feature of the island morphology is the development of a moderate-depth
131 carbonate shelf (~ 120 to 140 m depth) that becomes wider to the north / northeast of the
132 island (figure 4 in Le Friant et al., 2004).

133 There are important tectonic structures in and around Montserrat. The island has developed at
134 a major offset in a regional transtensional fault zone (Feuillet et al., 2010). To the SE of
135 Montserrat, the Bouillante-Montserrat Fault Zone (Fig. 1) bounds a major half-graben to the
136 east (Watt et al., 2012). The Havers-Montserrat fault system lies to the NW of Montserrat.

137 The same fault zone, known as the Belham Valley fault, extends across the island between
138 the SHV and Centre Hills along a WNW-ESE trend. The Belham Valley fault is a complex
139 zone of deformation with localised uplift of the St George's and Garibaldi Hills. Lava domes,
140 fumaroles and hot springs of the SHV are elongated along the fault zone (Wadge et al.,
141 2014). A major geothermal field is centred on the Belham Valley and St George's Hill to the
142 NW of the SHV (Ryan et al., 2013). Seismic sections across the submarine fault-zone
143 (Kenedi et al., 2010) show it is a major listric system downthrown to the NNE with multiple
144 minor faulting across the footwall side with localised uplift in the hanging wall.

145

146 *2.2 Kick'em Jenny volcanic field*

147 Kick'em Jenny submarine volcano (KeJ) is located just 7.5 km north of the island of Grenada
148 in the southern Lesser Antilles (KeJ, Fig. 2). The first detailed bathymetric survey of the
149 volcano in 1972 revealed a 1300 m high conical structure, constructed on the western flank of
150 the arc with a summit crater at 190 m water-depth (Sigurdsson and Shepherd, 1974). Recent
151 SEABEAM mapping of the volcano has shown that the summit cone and crater actually lie
152 within a much larger arcuate collapse structure that opens to the west (Sigurdsson et al.,
153 2006). A debris avalanche deposit associated with formation of the horseshoe-shaped
154 collapse structure extends 17 km downslope into the back-arc Grenada Basin (Lindsay et al.,
155 2005; Dondin et al., 2012) with chemosynthetic cold seeps at the distal end (Carey et al.,
156 2014a). KeJ has erupted at least 12 times since 1939 and is currently the most active volcano
157 in the West Indies (Devas, 1974; Lindsay et al., 2005). Some of the eruptions produce surface
158 disturbances, subaerial plumes and minor tsunamis, whereas others have been detected only
159 by T-phase seismic signals (Shepherd and Robson, 1967; Lindsay et al., 2005). Recent
160 multibeam mapping and remotely-operated vehicle explorations of the area have revealed the
161 presence of several other small volcanic centres just to the east and southeast of KeJ (Carey et

162 al., 2015). The slopes of these centres, lying at about 200 metres water depth and apparently
163 not recently active, exhibit common outcrops of carbonate crusts similar to those seen off the
164 coast of Montserrat.

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166 *2.3 Kolumbo Submarine Volcanic Line*

167 The seafloor northeast of Santorini volcano in Greece consists of a small, elongate rifted
168 basin that has been the site of recent submarine volcanism (Kolumbo, Fig. 2). This area lies
169 within the Cyclades back-arc region of the present-day Hellenic subduction zone where the
170 seafloor of the eastern Mediterranean Sea is descending beneath the Aegean microplate. The
171 Cycladic region and the Aegean Sea as a whole are known to be regions of south-easterly
172 back-arc extension and thinning of continental crust. Nineteen submarine volcanic cones
173 occur within this small rift zone, the largest of these being Kolumbo which last erupted
174 explosively in 1650 AD, causing significant damage and fatalities on the nearby island of
175 Santorini (Carey et al., 2011; Nomikou et al., 2012). Water depths of the submarine cones
176 range from 18 to 450 metres. In general, the domes and craters northeast of Kolumbo are
177 sediment covered and show little evidence of recent volcanic activity. Outcrops of volcanic
178 rock can be found in the crater walls and slopes of some of the cones, but they typically
179 consist of volcanic fragments of pumice and lava, along with some bioclastic material,
180 cemented by calcite, indicative of the lack of recent eruptions.

181

182 **3. Methods and Samples**

183 Multibeam mapping and remotely-operated vehicle (ROV) explorations of the submarine
184 flanks of Montserrat, Kick'em Jenny and the Kolumbo seamount chain in the Mediterranean
185 enabled submarine hardground occurrences to be imaged and sampled (Carey et al., 2011;
186 Carey et al., 2014a,b; Watt et al., 2015; Carey et al., 2015). In addition to *in-situ* samples in

187 the submarine environment, limestones have been sampled from three other settings on and
188 around Montserrat. The first is from outcrops of submarine volcanoclastic sediment with thin
189 (1-2 m thick) limestones on land at Roche's Bluff on the southeastern flank of Montserrat; a
190 volcanic clast here has an $^{40}\text{Ar}/^{39}\text{Ar}$ age of 1.02 Ma (Harford et al., 2002). Second,
191 geothermal boreholes in the Belham Valley have recovered limestones interbedded with
192 volcanics and intrusives in continuous core and in cuttings to depths of 2.3 km. And finally,
193 Holocene offshore debris-avalanche deposits contain clasts of shallow-water coralgall
194 limestone that have been transported into deeper water. Locations, water depths and brief
195 descriptions of the samples are given in Table 1.

196 All limestone samples, including those from the geothermal borehole and Roche's
197 Bluff outcrops, have been examined in thin-section. For stable isotope analysis of bulk
198 carbonate the sample material was ground in agate and 10 mg of carbonate powder was
199 reacted with anhydrous phosphoric acid in vacuo for 72 hours at a constant 25°C (Ca and Mg
200 carbonates including dolomite should react within this time) at the British Geological Survey
201 (BGS). The CO_2 liberated was separated from water vapour under vacuum and collected for
202 analysis. Measurements were made on a VG Optima mass spectrometer at BGS Keyworth,
203 UK. Overall analytical reproducibility for these samples is normally better than 0.2‰ for δ
204 ^{13}C and $\delta^{18}\text{O}$ (2σ). Isotope values ($\delta^{13}\text{C}$, $\delta^{18}\text{O}$) are reported as per mil (‰) deviations of the
205 isotopic ratios ($^{13}\text{C}/^{12}\text{C}$, $^{18}\text{O}/^{16}\text{O}$) calculated to the VPDB scale using a within-run laboratory
206 standard calibrated against NBS standards (Table 2).

207 Samples selected for strontium isotope seawater-age determinations were measured in the
208 Bristol Isotope Group facility at the University of Bristol. Following the protocol described in
209 Lewis et al. (2014), samples were dissolved in high purity acids and Sr separated using Sr
210 Spec resin (Horwitz et al., 1992). Samples were loaded on to single rhenium filaments with a
211 TaCl_5 activator (Birck, 1986) and Sr isotope ratios were measured using a Thermo-Finnigan

212 Triton thermal ionization mass spectrometer. Samples were measured using a multi-dynamic
213 ‘triple-jump’ method (Thirlwall, 1991; Avanzinelli et al., 2004) of 200 cycles with a 4.194 s
214 integration time per cycle. Instrumental mass bias was corrected using an exponential mass
215 bias law and an $^{86}\text{Sr}/^{88}\text{Sr}$ of 0.1194 (Nier, 1938; Russell et al., 1978); ^{87}Rb is corrected by
216 monitoring ^{85}Rb and using an $^{85}\text{Rb}/^{87}\text{Rb}$ of 2.59265 (Steiger and Jäger, 1977). Long-term
217 reproducibility of $^{87}\text{Sr}/^{86}\text{Sr}$ for NIST SRM 987 processed through column chemistry is
218 0.710247 ± 0.00001 (2 SD, $n = 58$). Results are given in Table 3.

219

220 **4. Results**

221 *4.1 Petrography of Carbonate Crusts*

222 *4.1.1 Montserrat Submarine Slope Limestones*

223 Multibeam images of the western and south-western flanks of the SHV are shown in Fig. 3. A
224 limestone crust occurs as a continuous layer across the submarine surface to water depths of
225 several hundred metres, as observed and videoed by the ROV. The crust shows abrupt
226 boundaries where large slabs have broken off and detached, indicating mass wasting. Figures
227 4a and 5a show images of limestone slabs where they have been locally disrupted and tilted,
228 revealing their internal texture and lithology. The limestone crusts are typically made of
229 volcanoclastic material (boulder to silt grade), containing variable amounts of bioclastic
230 debris cemented by carbonate. There are also crusts largely composed of carbonate skeletal
231 debris with subsidiary volcanic clasts. These are strongly lithified rocks and contrast with the
232 unconsolidated and uncemented volcanoclastic deposits that form when lahars and pyroclastic
233 flows enter the ocean from the island (Le Friant et al., 2009; Trofimovs et al., 2012).
234 Samples of cemented slope limestone were collected by ROV from the nearshore area to the
235 SW of Montserrat. Two samples were located some 1-2 km from the shoreline (Fig. 3), at
236 188 m water depth (NA037-029) and 292 m (NA037-033). Both came from seafloor

237 exposures of disrupted limestone crust (Figs. 4 and 5). Two other limestone samples were
238 located about 8 km from the shoreline (Figs. 6 and 7) and are associated with the debris
239 avalanche known as deposit 5 (Le Friant et al., 2004), formed between 8 and 12 kyr (Cassidy
240 et al., 2013) due to collapse of the shallow shelf platform. Sample NA037-025 comes from
241 806 m water depth and NA037-026 from 823 m.

242 Sample NA037-029 is a bioclastic volcanic-grain pack-grainstone with some pebble-sized
243 volcanic clasts (Fig. 4a, b). Some granules-pebbles are coated by calcareous red algae (giving
244 rhodoliths)(Fig. 4b); the sandy (< 2 mm) sediment consists of fragments of red and green
245 calcareous algae (including *Halimeda*), foraminifera (several *Amphistegina*), bivalve,
246 serpulid and echinoid. The sediment is moderately-well sorted with some larger coated
247 grains. There are sponge borings in shells (Fig. 4c), filled with sediment, but these borings
248 also penetrate the sediment, indicating an earlier phase of cementation. The micritic sediment
249 contains diffuse peloids, and possible filaments, probably of microbial origin. In areas of
250 intergranular pores and cavities, a fibrous-bladed isopachous calcite fringe occurs around
251 grains (Fig. 4d). This rock was likely deposited from a sediment gravity flow or landslide and
252 has been cemented *in situ* by a micritic and fibrous calcite cement to form a hardground.

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254 Sample NA037-033 is also a true hardground from syndepositional cementation of the
255 submarine slope (Fig. 5a). It is a bioclastic volcanoclastic grainstone (Figure 5b), with larger
256 clasts up to several cm, consisting of numerous shallow-water bioclasts: foraminifera
257 (*Amphistegina*, *Peneroplis*, miliolinids), bivalves, gastropods, echinoids, calcareous algae –
258 both red and green (*Halimeda*), and volcanic grains coated with calcareous red algae,
259 serpulids and encrusting foraminifera (Figs. 5b-d). Planktic gastropods (pteropods and
260 heteropods) are present as well as planktic foraminifera. Peloids, probably microbial, occur
261 within cavities in a geopetal arrangement (Fig. 5d). The main cement between grains consists

262 of either an isopachous bladed calcite fringe or an isopachous micritic fringe (Figs. 5e, f).
263 Within gastropods there are acicular aragonite cements, syntaxial upon the shell. There is still
264 much empty pore space. This deposit is interpreted as re-sedimented, consisting of a mixture
265 of volcanic crystals and clasts and shallow-water skeletal debris. However, there is also a
266 planktic component indicating some mixing with pelagic sediment during slope transport.
267 Sample NA037-025 is a loose piece of cemented sand (Fig. 6a) from a deep-water slope
268 within an area of surface crust. The sediment is extremely well sorted with all grains being
269 medium to coarse sand-size. There is a high content (90%) of bioclasts: shallow-water
270 foraminifera (peneropliids), calcareous algae (branched forms), green algae (*Halimeda*,
271 aragonitic, some with evidence of dissolution) and bivalve fragments (Fig. 6b). Volcanic
272 grains are a minor content (10%). Peloids are present in cavities between grains and are
273 probably microbial in origin. The cement is a bladed isopachous calcite, with polygonal
274 junctions between fringes, typical of marine phreatic precipitation (Figs. 6c, d). Acicular
275 aragonite cement occurs within gastropod shells, a syntaxial effect (Fig. 6d).
276 Sample NA037-026 is a loose sample from an area of rocky outcrops on a quite steep slope
277 (Fig. 7a). It consists of relatively well-sorted granule-small pebble sized volcanic clasts and
278 crystals, many coated with crusts of calcareous red algae (*Amphiroa*, *Lithoporella*),
279 foraminifera (including nubecularids and acervulinids), serpulids, bryozoans and microbial
280 laminae (Figs. 7b-d). Calcimicrobe filaments are present (like *Girvanella*), with some being
281 beaded (like *Rothpletzella*, Rob Riding pers. comm.). Pockets of phosphatic peloids are likely
282 also microbial-bacterial. Other bioclasts present include benthic foraminifera (*Amphistegina*
283 and *Archaias*) and echinoids. Evidence of dissolution is present in the form of bivalve and
284 heteropod-gastropod molds (Fig. 7e), some filled with calcite spar. Sponge spicules are also
285 present and show dissolution and replacement by calcite. The patchy cement is mostly a
286 sparry calcite with a drusy fabric. The benthic foraminifera are both from long-ranging

287 genera spanning the Eocene to Holocene and Middle Eocene to Holocene (Boudagher-Fadel,
288 2008) and the random orientation of sections prevents further identification to species level.
289 For both samples 025 and 026 the bioclastic and coated volcanic grain sediment must have
290 formed in much shallower water but then been transported to these deeper locations by either
291 a sediment gravity flow or submarine landslide, where the induration took place. Some
292 deeper-water bioclasts are also present (the heteropods, planktic foraminifera and sponge
293 spicules), probably picked up en-route.

294 In some cases, the cemented slope surfaces are coated with a thin layer of black Mn-Fe
295 oxide-hydroxide, which also occurs on submarine outcrops of volcanic rock. This distinctive
296 surface usually has a very irregular topography of 1-2 cm which likely reflects the effects of
297 boring organisms and local dissolution. Such Fe-Mn oxide coatings are typical of deeper-
298 water lithified surfaces and rock outcrops in areas of sediment starvation and/or where there
299 are quite strong currents sweeping the seafloor. Phosphate (honey-brown colour in thin-
300 section) may also be present, and commonly planktic foraminifera are trapped within the
301 coating. The origin of these Mn-Fe surfaces has been much debated but a bacterial origin is
302 suspected in many cases (Konhauser, 1998). Indeed, tufts of *Frutexitis* bacteria have been
303 observed in thin-section of one such coating.

304

305 *4.1.2 Montserrat Submarine Debris Avalanche Deposits*

306 Submarine debris avalanche deposits are a prominent feature of the seafloor around
307 Montserrat (Le Friant et al., 2004; Cassidy et al., 2013; Watt et al., 2015). Off the east coast
308 the youngest debris avalanche, deposit 1, is attributed to the formation of English's Crater by
309 sector collapse at about 4 kyr BP. Three samples of limestone were collected from deposit 1
310 (Fig. 2). Sample NA037-005 (depth 942 m and 12 km offshore) consists of a grey-green
311 micritic pelagic limestone with numerous planktic foraminifera (*Globorotalia*, *Orbicula*),

312 pteropods, thin-shelled planktic gastropods (heteropods), minor fragments of shallow-water
313 bioclasts (bivalve, foraminifera, echinoid), and silt-sized volcanic crystals. There are some
314 mm-size burrow structures. Samples NA037-002 (975 m depth and 12 km offshore) and
315 NA037-012 (840 m depth and 6 km offshore) are of a similar shallow-water microfacies.
316 Sample NA037-002 is a highly indurated carbonate with coralline algal fragments and small
317 volcanic lithics. Large (cm-size) rhodoliths (nodules of calcareous red algae) occur in a
318 matrix of micrite with many bioclasts, including bivalves, gastropods and benthic
319 foraminifera (*Amphistegina*, *Archaias* and peneroplids). Patches of drusy calcite spar occur in
320 cavities and occupy moulds of bivalves. Sponge borings occur in the rhodoliths, some filled
321 with microbial peloids. Sample NA037-012 is also coralgal limestone with rhodoliths and a
322 variety of bioclasts including peneroplid foraminifera and bivalves, some showing effects of
323 dissolution and replacement of aragonite by calcite. Encrusting forams and peyssoneliaceae
324 algae are present. Volcanic material is subordinate. Samples 002 and 012 are both shallow-to-
325 moderate (<50 m) depth limestones with typical lagoonal forams. The diagenetic features are
326 consistent with meteoric / freshwater alteration (Tucker and Wright, 1990), although
327 meteoric-type cements can form in deeper water (Schlager and James, 1978; Melim et al.,
328 1995, 2002). Overall, these samples represent limestones from the shallow shelf dislodged
329 and incorporated into the debris avalanche. Sample 002 contains a single probable
330 miogypsinid fragment, which would give a Miocene age; the fragmentary nature of the
331 bioclast however means that reworking is a possibility, although this does agree with the Sr
332 data. However, other genera present again have long ranges from Eocene to Holocene.

333 *4.1.3 Roche's Bluff volcanoclastics, Montserrat*

334 Limestones within a package of submarine volcanoclastics at Roche's Bluff in the SE corner
335 of Montserrat are much fractured and deformed, but fabrics are quite well preserved in thin-
336 section, with some patchy recrystallisation. MacGregor (1938) identified corals, echinoid

337 spines, serpulids, bivalves and ostracods in the limestones. In our samples the rock is a
338 corallgal limestone with fragments of volcanic rock, many coated with coralline algae and
339 encrusting foraminifera (including nubecularids). Gastropods and bivalves are present
340 (aragonite shells replaced by calcite), and benthic foraminifera: miliolids, *Amphistegina*,
341 poorly preserved *Archaias*-type forms, miogypsinids, rare planktic foraminifera and
342 agglutinating textularid foraminifera. There are two generations of calcite cement: fibrous
343 fringes and drusy spar, and minor aragonite cement has been replaced by calcite. This
344 limestone was deposited in a shallow-moderate depth, relatively high-energy location. It was
345 subjected to marine cementation and also meteoric dissolution-cementation. The presence of
346 multiple sub-axial sections of *Miogypsina* indicates an early Miocene age in the Americas
347 (BouDagher-Fadel, 2008); the specimens do appear to be complete.

348

349 *4.1.4 Montserrat Geothermal Wells*

350 Exploration for geothermal energy on Montserrat included three drill-sites. Continuous core
351 was collected in Well Mon3 at depths of 1.5, 1.7 and 2.0 km and rock cuttings were
352 recovered at all levels. Of most relevance here are two horizons of limestone at 2014-2016 m
353 and 930-1020 m depth, within a succession dominated by volcanic, volcanoclastic and
354 intrusive rocks. The limestones at 2014-2016 m, showing evidence of recrystallisation, are
355 mostly fossiliferous wackestone to pack-grainstone from shallow to moderate depths, normal
356 open-marine water. From the foraminifera, a late Miocene age has been assigned (Marcelle
357 BouDagher-Fadel, personal communication). Limestones from depths 1020 to 950 m are also
358 highly fossiliferous and indicate an upward trend from shallower to deeper water. Sample 930
359 m contains abundant planktic foraminifera, including likely *Orbulina universa*, a single
360 potential *Globergerinoides sacculifer* and potential *Globorotalia* with *Trilobatus trilobus*
361 indicating a likely age in the last 5 Ma.

362

363 *4.1.5 Kick'em Jenny, Southern Lesser Antilles*

364 Two samples are highlighted from the area of Kick'em Jenny submarine volcano in the
365 southern Lesser Antilles (Fig. 2). The first, NA039-071, is a partially indurated carbonate
366 found in proximity to a cold-seep area associated with the distal end of a large debris
367 avalanche on the western slope of KeJ at a depth of 1956 m (Figs. 2, 8a). The seeps host a
368 chemosynthetic-based ecosystem with bivalves, tubeworms, gastropods, crabs and
369 holothurians (Carey et al., 2014a, b, c). The hand-sample contains large burrows, up to 20
370 mm in diameter, some of which are open whereas others contain intraclasts and pellets, up to
371 7 mm in diameter (Fig. 8b, c). In thin-section the sediment consists of grey-green pelagic
372 lime mud and silt with many tiny microfossils including planktic forams (100-500 microns
373 across) and fine skeletal fragments (<200 microns). However, conspicuous are relatively
374 large ostracods, up to 3 mm across (Figs. 8c, d). Some of these ostracods with a mud-fill
375 appear to be part of the large pellets within burrows. Isopachous fibrous-bladed calcite
376 cement occurs locally around the pellets (Fig. 8d). Within one ostracod an acicular cement is
377 present, appearing more like aragonite. Of note, as reported later, this sample has an
378 extremely low $\delta^{13}\text{C}$ of -42‰.

379 The second sample, NA039-035, is from the upper slopes of the Kick'em Jack submarine
380 volcano at 172 m depth (Fig. 2) and consists of a mixture of volcanic grains, bioclastic
381 material and lime mud (Fig. 9a). Mafic crystals commonly have a reaction rim. The matrix
382 consists of silt-grade volcanic particles and lime mud which appears to have infiltrated
383 cavities between grains. This mud is vaguely peloidal, but locally it grades into a clear
384 micropeloidal texture. There are pockets of micropeloids, with no matrix between, occurring
385 within cavities which are lined by an initial, very thin isopachous honey-brown amorphous
386 material (Fig. 9b). These micropeloids, 10-30 μm in diameter, appear to have formed within

387 the cavities rather than being washed in, suggesting an in-situ microbial origin. There are also
388 larger peloidal structures (30-60 μm), some coalesced, composed of phosphate. In the
389 patches of lime mud, there are planktic forams, fine bioclastic debris and sponge spicules,
390 replaced by calcite; spicules also occur within empty cavities, which may well have been
391 sponge borings.

392

393 *4.1.6 Kolumbo Seamount Chain, Mediterranean*

394 The numerous seamounts extending northeast from the Santorini caldera show little
395 evidence of recent volcanic activity, with the exception of Kolumbo, which is mostly
396 sediment covered with local outcrops of volcanic rock (Nomikou et al., 2012). Many of the
397 slopes of the seamounts consist of brown, carbonate crusts that are commonly fractured into
398 slab-like pieces. The crusts are typically tens of centimetres to a few metres in thickness with
399 a thin draping of unconsolidated hemipelagic sediment. The sampling locations of three
400 representative samples of carbonate crust from this area are shown in Fig. 2. Samples
401 NA007-001 and NA007-045 are from a small dome complex on the northeast margin of the
402 Kolumbo submarine volcano in water depths of 200 and 128 metres respectively (Fig. 2).
403 Sample NA007-022 was collected from a depth of 194 m on a separate small seamount about
404 7 km northeast of Kolumbo.

405 Sample NA007-001 is a pelagic limestone with many microfossils, planktic forams, thin-
406 shelled bivalves as well as fragments of sponge spicules. The sample itself shows the effects
407 of boring sponges (Fig. 10a). Indeed, an earlier phase of boring has cavities filled with a more
408 brownish lime mud compared to the host lime mudstone that has a more grey colour. Later
409 sponge borings are open and there are modern encrusting serpulids and other organisms
410 present too. Sample NA007-022 (Fig. 10b) has granule to pebble-sized volcanic clasts coated
411 by calcareous red algae, with lime mud containing microfossils between. The sample has also

412 been bored by sponges and cavities are in places lined by a bladed calcite fringe before a later
413 phase of more microfossiliferous lime mud filling cavities (Figs. 10c, d).

414 Sample NA007-045 is from slightly deeper water (128 m), on the side of a very small
415 volcanic dome (Figs. 2, 11a). This is a crustose limestone composed of an open framework of
416 foliaceous coralline algae, encrusted with bryozoans and serpulids, with fine-medium sand
417 and volcanic-bioclastic material occurring within the framework cavities (Figs. 11b-d).

418 Fibrous calcite cements occur here as crusts and fans.

419 These samples from the Hellenic Arc in the Mediterranean are similar to the crusts from the
420 Caribbean: a variable mixture of volcanic and bioclastic material, the biota depending on
421 water depth and degree of downslope reworking of skeletal grains; early cementation
422 commonly by fibrous calcite and micritic calcite; several phases of sponge boring with lime
423 mud, usually pelagic, filling cavities and latest borings still empty, and crusts in deeper water
424 showing thin Fe-Mn coatings.

425

426 *4.2 Isotope data*

427 Oxygen, carbon and strontium isotopic compositions of limestone samples are presented in
428 Tables 2 and 3. Apparent and approximate ages of carbonate samples can be estimated based
429 on strontium isotopic ratios in comparison with the global seawater strontium isotope curve
430 and data from Neogene carbonates (e.g. Farrell et al., 1995; Howarth and McArthur, 1997 –
431 LOWESS Database 2013; McArthur et al., 2006; McArthur et al., 2012). This approach
432 assumes that the carbonate minerals have been precipitated from seawater and that they have
433 not subsequently been perturbed. However, some of the limestones, notably those from the
434 geothermal well, have ages that are apparently much older than those indicated by
435 foraminifera, suggesting that their initial Sr isotopic compositions have been altered; this is

436 interpreted to have been through contact with geothermal fluids as discussed in more detail
437 below.

438 Figure 12 shows a plot of the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ isotope data. Samples of slope crusts NA037-033
439 and NA037-029 from nearshore SW Montserrat have young Sr isotope ages of less than 200
440 kyr and very similar $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$, averaging +3.6 ‰ and 2.25 ‰ respectively. The $\delta^{13}\text{C}$ are
441 within the usual range of marine sediments (Tucker and Wright, 1990), whereas the $\delta^{18}\text{O}$ is a
442 little more positive than might be expected for a tropical carbonate (e.g. Swart, 2015),
443 possibly since the sediment was cemented in water > hundred metres deep, which would have
444 been quite cool (~13-20 °C), compared with a near-surface water temperature of 28-30 °C
445 (Kameo et al., 2004). Sample NA037-025 from offshore SW Montserrat also has a young Sr
446 isotope age (a few 100 kyr) and similar marine values for both $\delta^{13}\text{C}$ (+3.2‰) and $\delta^{18}\text{O}$
447 (+2.5‰). In contrast, sample NA037-026 has a much older apparent Sr isotope age (17-20
448 Ma) and low negative stable isotope values of both $\delta^{13}\text{C}$ of -2.3‰ and $\delta^{18}\text{O}$ -3.8‰,
449 suggesting alteration.

450 The three limestone clasts collected from Montserrat debris-avalanche Deposit 1 on the
451 eastern side of the island have diverse isotope geochemistry. The pelagic limestone (sample
452 NA037-005) has a very young Sr isotope age (<100 kyr) and typical marine values for $\delta^{13}\text{C}$
453 and $\delta^{18}\text{O}$. The two coralline limestone clasts (NA037-002 and NA037-012) have yielded
454 much older Sr age dates, especially sample NA037-002, of 23-25 Ma (early Miocene),
455 whereas sample NA037-012 is 975 kyr to 1.5 Ma (Table 3). For the C and O isotopes,
456 sample NA037-012 has low positive values (Fig. 12), both likely marine. Sample NA037-
457 002, on the other hand, has a more typical marine $\delta^{13}\text{C}$ signature (+2‰) but $\delta^{18}\text{O}$ varies, with
458 4 negative values (-2.5 to -3.6‰) and one of +2‰. The negative $\delta^{18}\text{O}$ values are consistent
459 with some alteration, likely meteoric, reflecting the low negative isotopic composition of
460 freshwater (mean rainfall is ~-4‰ $\delta^{18}\text{O}$ VSMOW for Barbados for example). The $\delta^{13}\text{C}$ values

461 indicate recycling of the original marine carbonate, rather than any effect of soil CO₂ gas
462 which would be enriched in (biogenic) ¹²C (Tucker and Wright, 1990).

463 Roche's Bluff limestone samples also yield apparent old Sr isotope ages: 16.5 and 17.1 Ma
464 (early Miocene), which are inconsistent with other age data, such as the ⁴⁰Ar/³⁹Ar age of
465 1.021 ±0.020 Ma from a volcanic clast in the Roche's Bluff Submarine Volcaniclastic unit
466 and the <1.8 Ma age suggested by the identification of the foraminifera species *Globorotalia*
467 *truncatulinoides* (Harford et al., 2002). However, the presence of several *Miogypsina* in the
468 thin-sections would support a Miocene age, although other larger foraminifera present only
469 indicate Eocene to Recent. The δ¹³C values are normal marine (average +2.1‰). However,
470 δ¹⁸O is very negative at -7.7‰; this is too low for rainfall of this latitude for it to be simply a
471 meteoric / freshwater effect. An alternative explanation is that the low negative δ¹⁸O of the
472 Roche's Bluff limestone was generated by recrystallisation (recorded in thin-section) under
473 moderate-high temperatures through interaction with hydrothermal fluids. This would also
474 account for the apparent old Sr ages, if the miogypsinids are reworked. Such hydrothermal
475 fluids, with a low Sr isotope value derived from passage through volcanics, may have been
476 introduced through the fault system adjacent to the Roche's Bluff outcrops.

477 For limestones in the geothermal well Mon3 the apparent Sr isotope ages are all much older
478 than the ages based on fossils and unpublished ⁴⁰Ar/³⁹Ar data. The core samples of upper
479 Miocene (~5-7 Ma) limestone at 2 km depth give much older apparent Sr ages in the range
480 16.7-19.7 Ma. Cuttings samples from 930 m and from 1198 m depth are also apparently old:
481 4 analyses give an age of around 15 Ma, although one analysis yielded an age of 0.55 Ma; the
482 planktic foraminifera do suggest a young age. Unpublished ⁴⁰Ar/³⁹Ar ages of andesites at
483 depths of 1.5 and 1.7 km depth are approximately 1 Ma old. The C and O isotope data from
484 two (whole-rock) limestone core samples both have negative δ¹³C and δ¹⁸O. For one sample
485 (2015 m depth) δ¹³C values (4 analyses) are low (average -0.75‰), with very negative δ¹⁸O

486 (average -10.3‰). For sample depth 2016 m, $\delta^{13}\text{C}$ values (2 analyses) are more negative
487 (average -3.2‰) and $\delta^{18}\text{O}$ is on average -9.4‰. One analysis from a thick-shelled bivalve in
488 that 2016 m sample has $\delta^{13}\text{C}$ of +1.9 and $\delta^{18}\text{O}$ of -11.2‰. The negative $\delta^{13}\text{C}$ values are likely
489 the result of decomposition of organic carbon within the sediments, producing isotopically
490 light CO_2 . For cuttings samples (930 m to 1198 m), $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ are around +2‰ for $\delta^{13}\text{C}$,
491 a typical marine carbonate value, but $\delta^{18}\text{O}$ is -8‰, again suggesting significant alteration
492 related to burial diagenesis / interactions with hydrothermal fluids although this had no effect
493 on the $\delta^{13}\text{C}$, possibly related to issues around supply or availability of C in the fluid.
494 These Sr and stable isotope analyses from the Mon3 well all indicate post-depositional
495 alteration of the original seawater-marine values, most likely by interaction with
496 hydrothermal-volcanic fluids. The apparent old ages are the result of introducing a
497 component of volcanic non-radiogenic Sr into the carbonates. $^{87}\text{Sr}/^{86}\text{Sr}$ of Montserrat
498 volcanic rocks are in the narrow range 0.7035 to 0.7038 (Cassidy et al., 2012). The Sr in the
499 geothermal fluids will be largely derived from the volcanic rocks and this will exchange with
500 or add to the limestones' initial Sr. Likewise, the strongly negative $\delta^{18}\text{O}$ reflects geothermal
501 fluid interactions. We surmise that the anomalously old Sr ages and negative $\delta^{18}\text{O}$ in samples
502 from the Deposit 5 debris avalanche (NA037-026), the Deposit 1 debris avalanche (NA037-
503 002) and the Roche's Bluff limestone, are also the consequence of hydrothermal alteration of
504 shelf limestones.

505

506 **5. Discussion**

507 We propose a conceptual model for the development of the limestone crusts based on
508 established groundwater models for coastal regions and volcanic islands (Cooper, 1959; Join
509 et al., 2005), perturbed by the development of a hydrothermal system (Fig. 13). Montserrat
510 broadly conforms to this conceptual model (Hemmings et al., 2015) of a freshwater lens

511 above sea level and saline (seawater) intrusion beneath with a transitional mixing-zone
512 occurring close to the coast. Freshwater outflows at the coast are balanced by recharge and
513 seawater inflow below sea level. The hydrogeology on Montserrat is significantly perturbed
514 due to the development of a geothermal field along the Belham Valley fault system (Ryan et
515 al., 2013) and the presence of an active hydrothermal system associated with the SHV
516 edifice. Several scattered fumaroles and hot springs have been present historically (Chiodini
517 et al., 1996), although these systems were disrupted by the 1995-2010 eruption. The Belham
518 Valley geothermal field contains brines with the approximate salinity of seawater, but with a
519 geochemistry suggestive of interaction with volcanic rocks. Temperatures up to 230 °C are
520 recorded in the geothermal wells.

521 We envisage that the carbonate crusts largely developed as a result of carbonate-saturated
522 seawater fluxing through the submarine flanks of the island, precipitating carbonate cement
523 in the pores of the sediments. The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values of many of the crust samples indicate
524 a seawater origin. In the classic coastal hydrological model (Fig. 13) the main driver is the
525 density difference between seawater and fresh groundwater in the volcanic edifice. The
526 subaerial flanks of the volcano are composed of high permeability and high porosity
527 volcanoclastic deposits which create the main aquifer (Hemmings et al., 2015). Two factors
528 can enhance the submarine inflow of seawater into the volcanic edifice. Upwelling of hot
529 geothermal and hydrothermal magmatic fluids within the volcanic island is expected to cause
530 inward flow of groundwater and underlying brines, strengthening the inflow of seawater. The
531 process of Kohout convection, driven by geothermal heat, has been invoked as a mechanism
532 for drawing seawater into carbonate shelves and isolated platforms, and precipitating
533 dolomite (Whitaker and Smart, 1990). In addition, inflow will be increased during periods of
534 sea-level rise due to the increased hydraulic head in the ocean (Tucker, 1993; Kim et al.,
535 2010); there have been changes in sea level on the scale of 100 m many times over the last

536 few million years as a result of the waxing and waning of Quaternary ice sheets. The inflow,
537 however, may decrease as the carbonate crust forms, with consequent reduction of porosity
538 and permeability, so that the formation of the crust may then promote sealing of the island
539 interior from seawater intrusion.

540 The formation of hardgrounds in the deeper-water slope environment from influxing seawater
541 is controlled by several factors, but notably carbonate saturation, the movement of fluid
542 through sediment (the ‘hydrodynamic level’ of Christ et al., 2015) and microbial activities, as
543 well as temperature and a sediment’s poroperm. At the depths of less than 1000 m where the
544 cemented crusts occur seawater will be saturated with respect to CaCO_3 , even though seafloor
545 temperatures will be down to 5°C (Kameo et al., 2004). The circulation of fluid through
546 sediment is essential to drive cement precipitation and here waves and tides, internal waves
547 and ocean currents impinging on a slope may all be involved (van der Kooij et al., 2010). In
548 addition, circulation driven by geothermal heat within the seamount, as discussed above, is
549 likely to contribute to drawing in water from the adjacent sea (Jones et al., 2000). With regard
550 to microbial influences, some bacteria promote carbonate precipitation, notably the
551 cyanobacteria and sulphate reducers, by raising alkalinity, whereas others, such as aerobic
552 heterotrophs and fermenters, may lead to local carbonate dissolution through reducing pH in
553 their metabolic processes (Dupraz et al., 2009).

554 Reviewing the diagenesis observed within the crusts, the most common cement is calcite, in
555 many cases a bladed-fibrous type, particularly as an isopachous rim around grains in the more
556 porous well-sorted, sand-grade deposits. Other crusts have a drusy sparry calcite cement
557 within cavities. Acicular aragonite crystals mostly occur as syntaxial precipitates on
558 originally aragonitic skeletal fragments, especially gastropods. Evidence of aragonite
559 dissolution and replacement is provided by molds of bivalves, some filled with calcite;
560 sponge spicules (originally opaline silica) have also been replaced by calcite, with crystals

561 extending out into cavities. Several phases of boring by sponges are evident as well as phases
562 of internal sedimentation. Micritic peloids are a feature of many crusts, occurring within
563 intraskeletal cavities, borings and between grains, and in some cases there is gradation into
564 micritic sediment. Microbial filaments are also present but they can be difficult to distinguish
565 from nubecularid foraminifera or thin poorly-preserved coralline algal crusts. Dissolution of
566 the surface of the hardgrounds is evident from their irregular microtopography and a mm-
567 thick layer of Mn-Fe oxide coating is a common feature. All in all, the paragenesis of these
568 crusts is commonly a complex series of cementation-alteration-boring-dissolution events.
569 All the features of the crusts are consistent with the volcanoclastic-carbonate sediment being
570 cemented and altered on the seafloor in the marine environment. They are similar to crusts
571 reported by Thompson et al. (1968), Schroeder et al. (2002), McKenzie and Bernoulli (1982)
572 and Buchs et al. (2018). The bioclastic material in the slope limestones described here is
573 dominated by calcareous algae, especially in the form of rhodoliths, encrusting volcanic
574 grains, and foraminifera, both benthic and planktic, the former commonly derived from
575 shallower water. Rhodoliths are a common feature of limestones associated with
576 volcanoclastic and volcanic rocks, especially in the moderate to high-energy settings of open-
577 ocean seamounts and islands (e.g. Madeira and the Azores; Baarli et al., 2014; Rebelo et al.,
578 2016). Calcite spar as a cement is usually regarded as a near-surface meteoric or burial
579 precipitate, but there are records of it occurring on the seafloor in deeper water (Melim et al.,
580 1995, 2002). Slower rates of precipitation in cooler water are the likely explanation. Cooler
581 water and its circulation through the surficial crusts are likely to account for aragonite
582 dissolution and replacement, again a feature commonly ascribed to freshwater diagenesis
583 (Tucker and Wright, 1990). Petrographic evidence for microbial activity is being recognized
584 more frequently as the significance of microbes in mineral precipitation is becoming
585 appreciated (e.g., Perri et al., 2018). Here in these crusts, communities of bacteria living

586 within the sediment are implicated by the occurrence of the peloidal micrite and it may well
587 be that microbial processes have contributed to the lithification of the slope sediment.

588 Most of the limestones in the geothermal wells, two clasts of shallow-water limestone taken
589 from debris-avalanche deposits, and the Roche's Bluff limestone, all show isotopic evidence
590 for hydrothermal alteration with very negative $\delta^{18}\text{O}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ compositions that indicate
591 ages from the seawater calibration curve that appear to be too old. We infer that these
592 limestones have been affected by the volcanogenic hydrothermal systems that developed
593 within the island interior. The observed alteration testifies to largescale hydrothermal
594 convection throughout the island's history.

595 The formation of these crusts is inferred to be in the mature stage of volcanic edifice growth.
596 Each of the volcanic centres initially formed on the seafloor at water depths of 800 to 1000 m
597 and grew to a height of the order of 1000 m above sea level, as is the case for the still active
598 Soufrière Hills Volcano. For a constant rate of volcanism the lateral speed of advance of the
599 volcanoes' flanks will slow; for example if the edifice is treated as a cone with a constant
600 height to width ratio then the lateral speed will decrease in proportion to $t^{-2/3}$.

601 There is geomorphological evidence for the stabilisation of the submarine flanks of
602 Montserrat and their preservation for long periods of time. Le Friant et al. (2004) compared
603 several topographic profiles around Montserrat from the shallow shelves to deep water. The
604 overall width of the shallow part of the island stays approximately the same from north to
605 south (Fig. 1) whereas the shallow shelf decreases in width. The profiles from the shelf edge
606 to deep water are very similar. These observations suggest that the submarine parts of the
607 islands stabilize after the volcano becomes extinct, which can be explained by the formation
608 of the limestone crust which prevents inward erosion by mass wasting. The widening of the
609 shallow shelf from south to north was interpreted by Le Friant et al. (2004) as the
610 consequence of lateral erosion of the island during glacial high stands due to wave and storm

611 erosion. However, there is new evidence for regional subsidence (Carey et al., 2019) and so
612 the widening of the platform to the north could also reflect subsidence of the three volcanic
613 centres on Montserrat, which become older to the north. In either case the platform outer
614 edge will approximately reflect the width of the final edifice at the cessation of volcanic
615 activity.

616 On the western side of Montserrat, there is further evidence for stabilisation of the submarine
617 flanks from the marked embayments and southward narrowing of the shelf. This zone is the
618 location of where active faults of the Havers-Montserrat fault system intersect the island. We
619 suggest that faulting in this zone has disrupted the limestone crust and promoted mass-
620 wasting of the submarine flanks.

621 Furthermore, episodes of sector collapse can play an important role in that they would
622 channelize lahars and pyroclastic flows down preferential pathways, as exemplified by the
623 Tar River Valley on the eastern flanks of Montserrat. As a consequence, large sections of
624 submarine coast will be abandoned with lateral growth reducing or ceasing, providing
625 conditions for thick limestone crust to form.

626 **6. Conclusions**

627 Remotely-operated vehicle (ROV) explorations of shallow to moderately deep seamounts in
628 many subduction zone environments, such as the Lesser Antilles (West Indies) and Hellenic
629 Arc (Greece), reveal the existence of extensive carbonate crusts on their submarine flanks.

630 These crusts are composed of mixtures of volcanoclastic debris and bioclastic material, in
631 most cases cemented by calcite, in the form of isopachous coatings around grains or pore-
632 filling spar. $^{87/86}\text{Sr}$ isotope data give an indication of age but also demonstrate that some
633 samples are altered by hydrothermal-volcanic processes, as shown by ages much older than
634 expected. Such alteration is also supported by $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ isotope data, although most
635 samples possess typical marine values, with the $\delta^{18}\text{O}$ commonly a little more positive, likely

636 reflecting cooler water at the depth of carbonate lithification. The occurrence of such crusts
637 on the slopes of seamounts and volcanic islands is significant from two perspectives. First,
638 they represent a new location of limestone deposition in the World's oceans. Second, the
639 hardgrounds act to protect the submarine flanks of volcanic islands and seamounts from
640 erosion and collapse and they may inhibit submarine intrusion of seawater into the interior of
641 such volcanic edifices. In addition, they also form important hard substrates for the
642 development of benthic biological communities.

643

644

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654

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976 **Captions to figures**

977

978 Fig. 1. Map showing the location of Montserrat island in the northern Lesser Antilles and the
979 bathymetry of the surrounding seafloor. Principal volcanic centers on Montserrat are marked

980 by red triangles. Faults are indicated based on Feuillet et al. (2010) and on interpretation of
981 seismic reflection data in Watt et al. (2012). The inset map shows the location of the study
982 region within the Lesser Antilles. Locations of seismic reflection lines from previous studies
983 are indicated by the blue and green lines that cross the Kahouanne valley.

984

985 Fig. 2. Maps showing the location of carbonate samples from a) Kick'em Jenny submarine
986 volcano area in the southern Lesser Antilles and b) Kolumbo submarine volcano and other
987 seamounts northeast of the Santorini volcanic complex in the southern Aegean Sea. Sample
988 localities are shown with red stars. Insert map in the Kolumbo figure shows the location of
989 the sampling area relative to Santorini.

990

991 Fig. 3. High-resolution multibeam map of the area southeast, south, and east of the island of
992 Montserrat. Carbonate samples used in the study are shown by red stars. Depth key is in the
993 lower right-hand corner of the figure. Irregular hummocky seafloor to the south and east of
994 Montserrat consists of large-scale debris avalanche deposits.

995

996 Fig. 4. Montserrat SW Nearshore. a) Collecting sample NA037-029 at 188 m depth from low
997 cliff. b) Well-indurated volcanoclastic-bioclastic pack-grain-rudstone with volcanic clasts
998 coated by coralline algae. c) Micrite-filled sponge borings cutting shell and adjacent sediment
999 indicating 2 phases cementation (thin-section, ppl). d) Grains cemented by isopachous fibrous
1000 calcite crust (thin-section, ppl).

1001

1002 Fig. 5. Montserrat SW Nearshore. a) Site of sample NA037-033 at 292 m. b) Cemented slope
1003 sediment consisting of volcanic clasts encrusted with calcareous algae, in a bioclastic

1004 volcanoclastic pack-grain-rudstone with shell fragments. c) Encrustation of volcanic clast
1005 with foraminifera, serpulids and calcareous red algae (thin-section, ppl). d) Peloids between
1006 grains, probably microbial (thin-section, ppl). e) Volcanic and bioclastic grains cemented by
1007 isopachous calcite fringe (thin-section, xp). f) Isopachous calcite cement fringe around
1008 bioclasts (thin-section, ppl).

1009

1010 Fig. 6. Montserrat SW Offshore, sample NA037-025, depth 806 m. a) Cemented bioclastic-
1011 volcanoclastic grainstone. b) Bioclasts, intragranular peloidal sediment, fringing calcite
1012 cement (thin-section, ppl). c) Isopachous fibrous calcite cement around grains (thin-section,
1013 ppl). d) SEM-BSE micrograph of calcite cement around bioclasts and aragonite cement
1014 within gastropod (upper left).

1015

1016 Fig. 7. Montserrat SW Offshore. a) Collecting sample NA037-026, depth 823 m. b), c)
1017 Bioclastic-volcanic pack-grain-rudstone with bio-coated volcanic grains (thin-section, ppl). d)
1018 Encrustation of volcanic clast by foraminifera, coralline algae, microbialite with filaments
1019 and serpulid (thin-section, ppl). e) Gastropod shell (formerly aragonite) dissolved out; whorl
1020 filled with calcite spar cement (thin-section, ppl).

1021

1022 Fig. 8. Kick'em Jenny, off Grenada. a) Sampling the seafloor crust. b) Sample NA039-071,
1023 1956 m depth. Pelagic lime mudstone with large burrows. c) Intraclasts and pellets of lime
1024 mud within burrow cavity. Patchy calcite cement fringes around clasts (thin-section, ppl). d)
1025 Ostracod filled by microfossiliferous lime mud; patchy bladed-fibrous calcite cement around
1026 shells (thin-section, ppl).

1027

1028 Fig. 9. Kick'em Jenny sample NA039-035, depth 172 m. a) Volcaniclastic material with
1029 patches of dark lime mud, some of which may have infiltrated cavities. Note cavities filled
1030 with peloids (thin-section, ppl). b) Close-up of micritic peloids, probably of microbial origin,
1031 within cavities (thin-section, ppl).

1032

1033 Fig. 10. Kolumbo seamount. a) Sample NA007-001 depth 200 m – a pelagic limestone with
1034 fine volcanic material, with 2 phases of sponge boring, 2nd phase empty, and bivalve shell
1035 (thin-section, ppl). b) Collecting sample NA007-022, depth 194 m, with several crust layers.
1036 c), d) NA007-022 A complex volcaniclastic limestone calcareous alga with cavities (likely
1037 sponge borings) lined by a thin calcite cement crust, followed by an influx of pelagic
1038 sediment. Empty sponge borings from a later phase of boring (thin-section, ppl).

1039

1040 Fig. 11. Kolumbo seamount. a) Collecting sample NA007-045, depth 128 m. b), c), d) Loose
1041 coralline red algal framework with fine volcaniclastic-bioclastic material partly filling growth
1042 cavities. (b), c) thin-section, ppl; d) (xp).

1043

1044 Fig. 12. Carbon and oxygen isotope cross-plot (‰ VPDB) for crust samples and for samples
1045 from the geothermal well Mon3 and Roche's Bluff. Sample names are given on the figure for
1046 values plotting outside the marine area (i.e. positive $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$). Also shown are the
1047 positions of the $\delta^{18}\text{O}$ values (but ‰ VSMOW) for geothermal and spring water from
1048 Hemmings et al. (2015) and Chiodini et al. (1996) respectively, and for fresh and altered
1049 SHV andesite from Zellmer et al. (2003).

1050

1051 Fig 13. Schematic model for formation of slope crusts on a seamount developed through

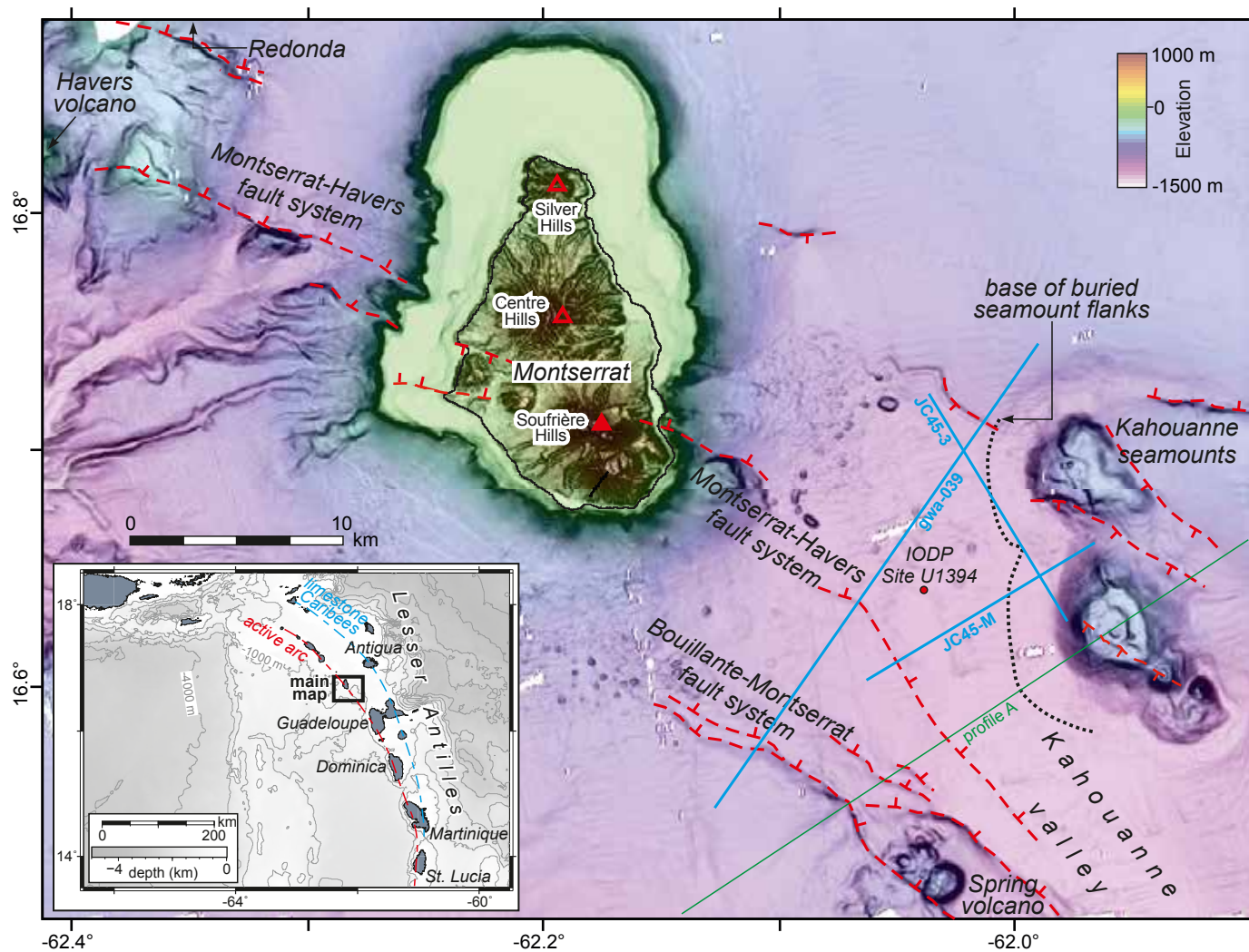
1052 precipitation of carbonate from seawater entering the volcanic edifice through ocean currents,
1053 waves and storms, and seawater drawn in through Kohout convection.

1054

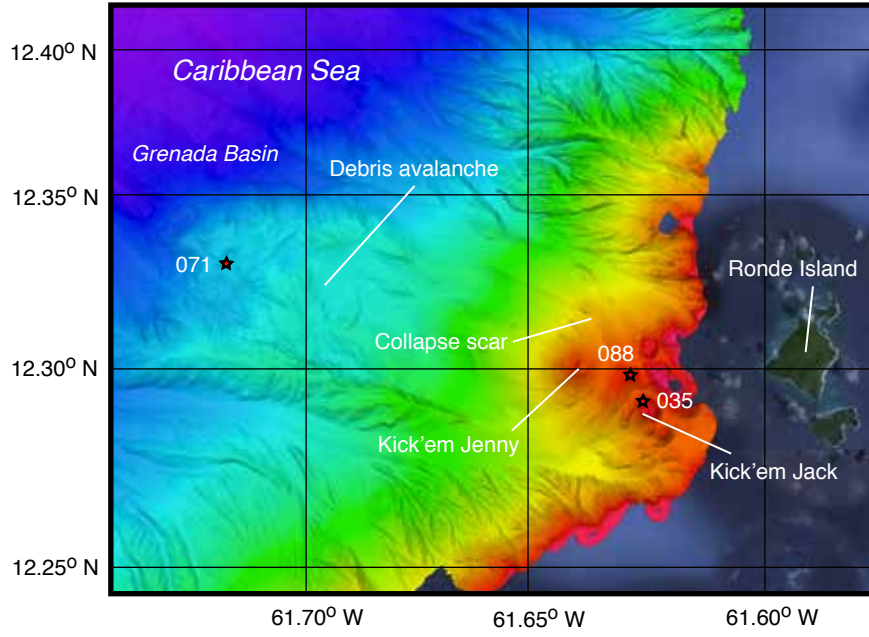
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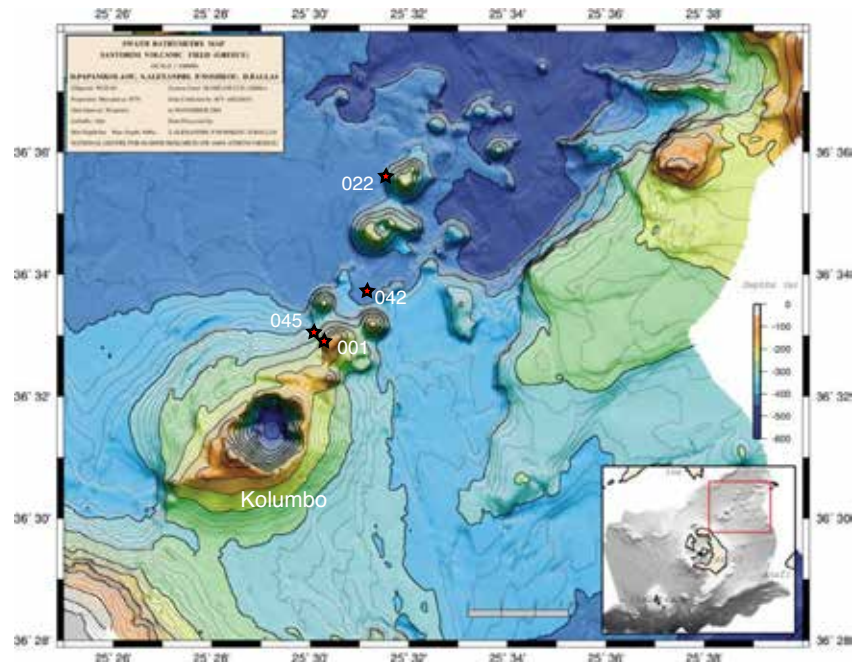
1057

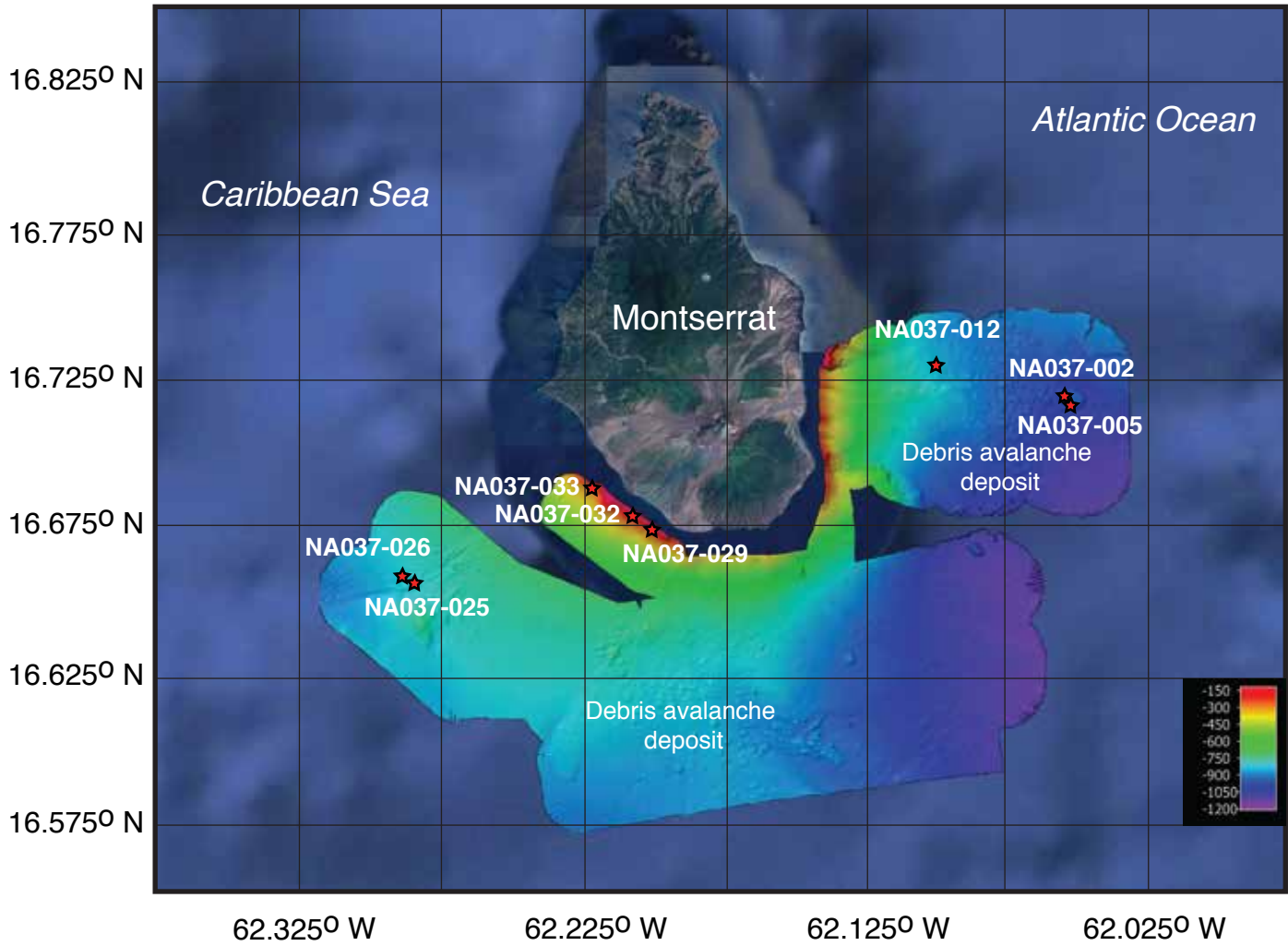


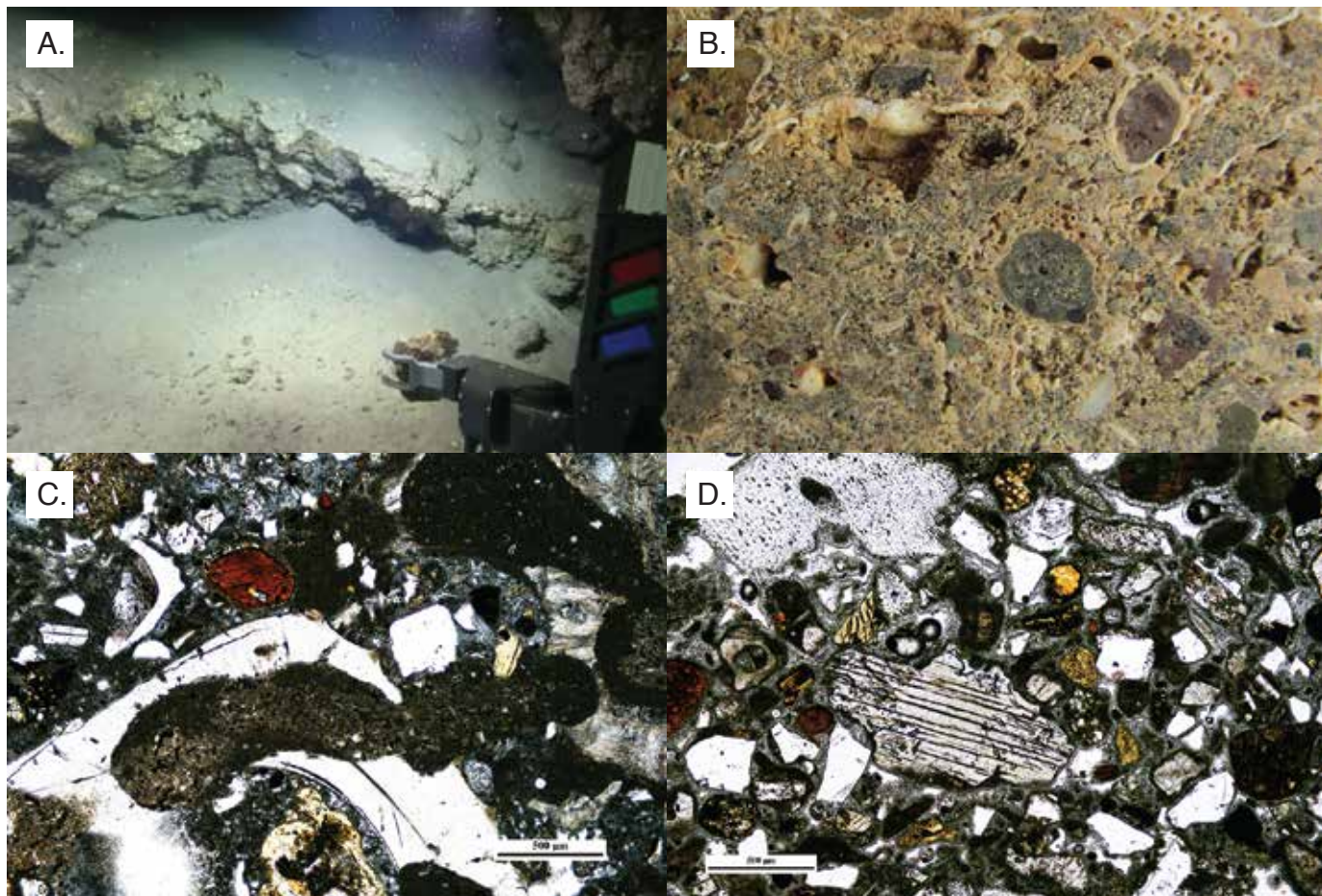
A.

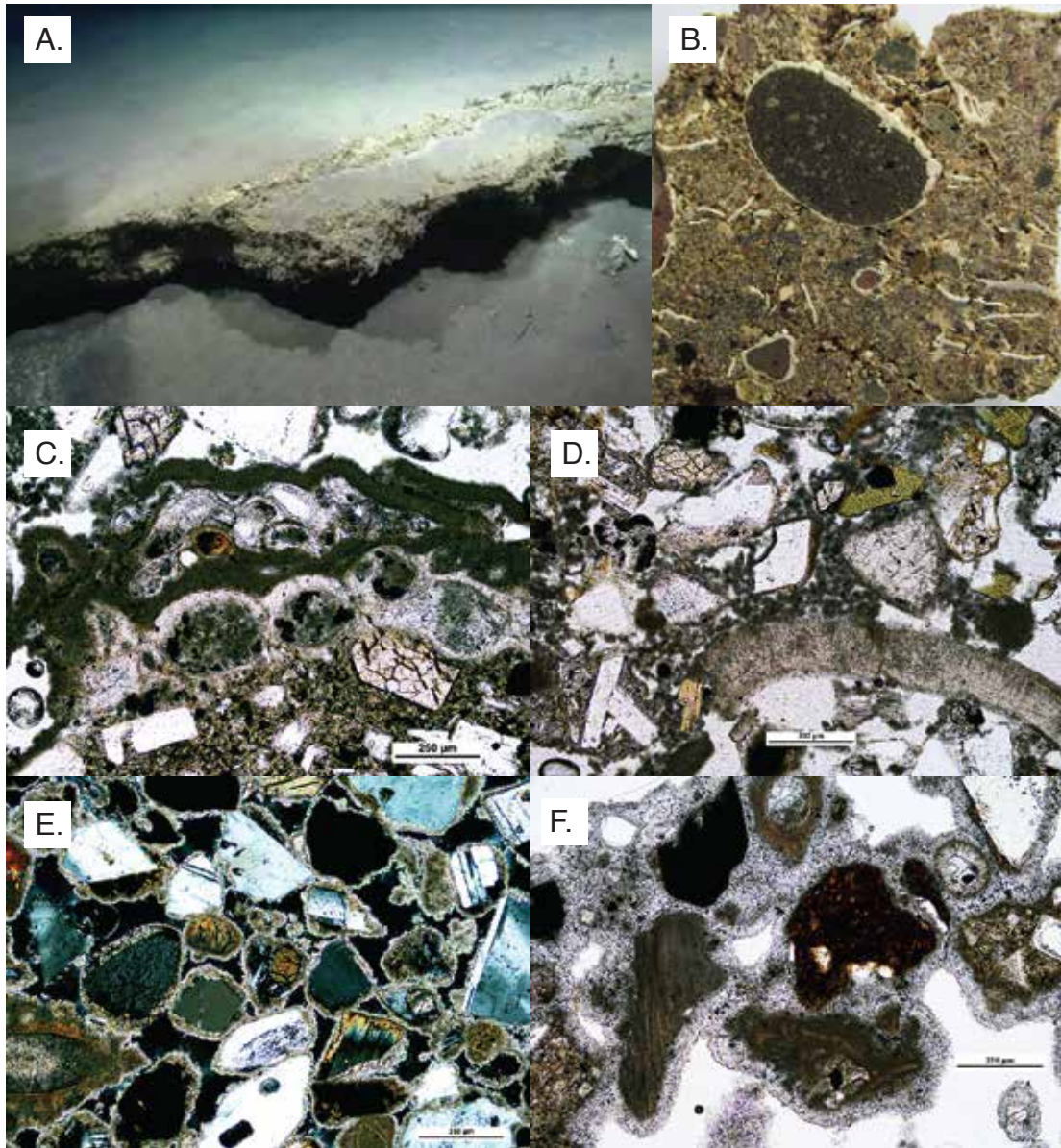


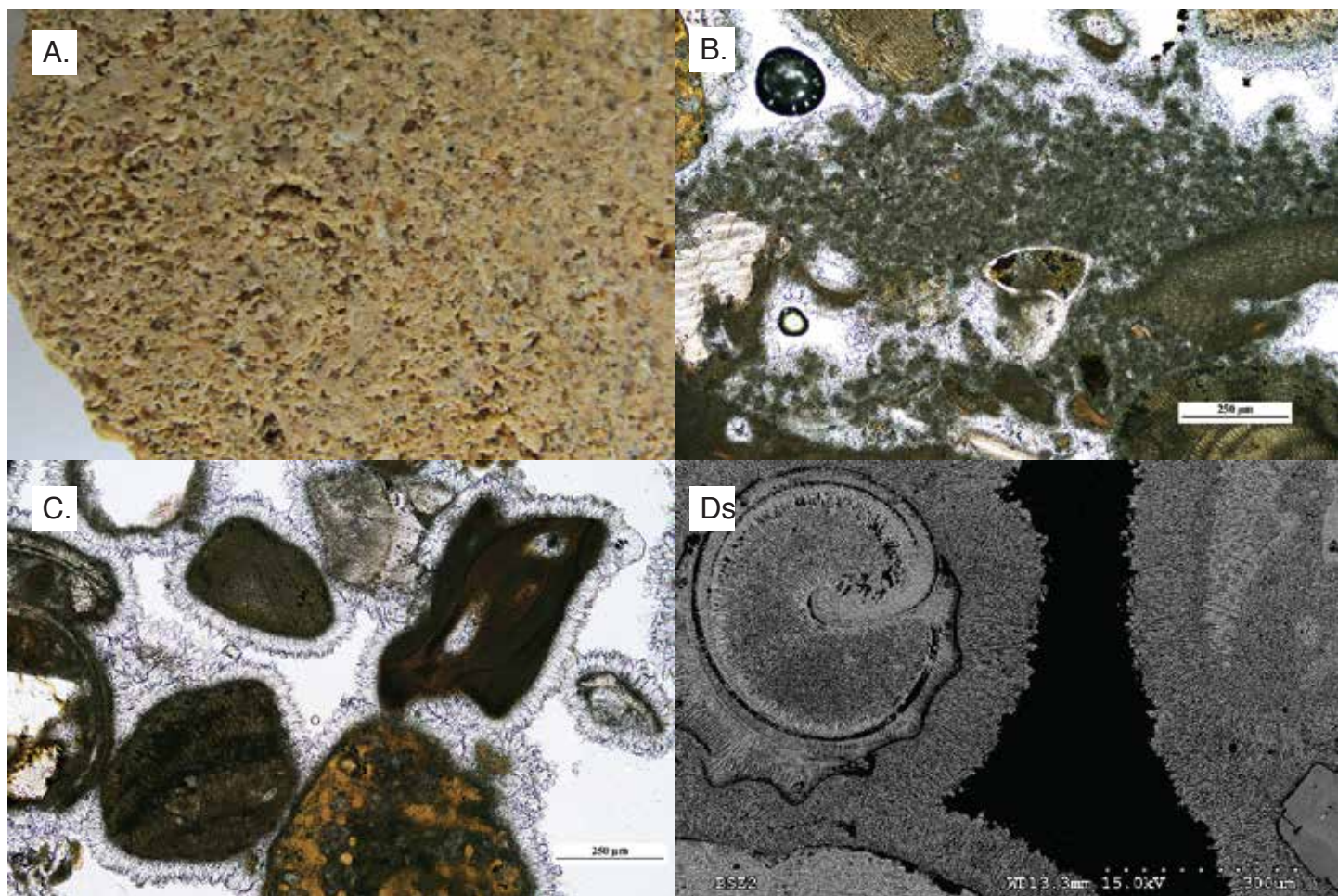
B.

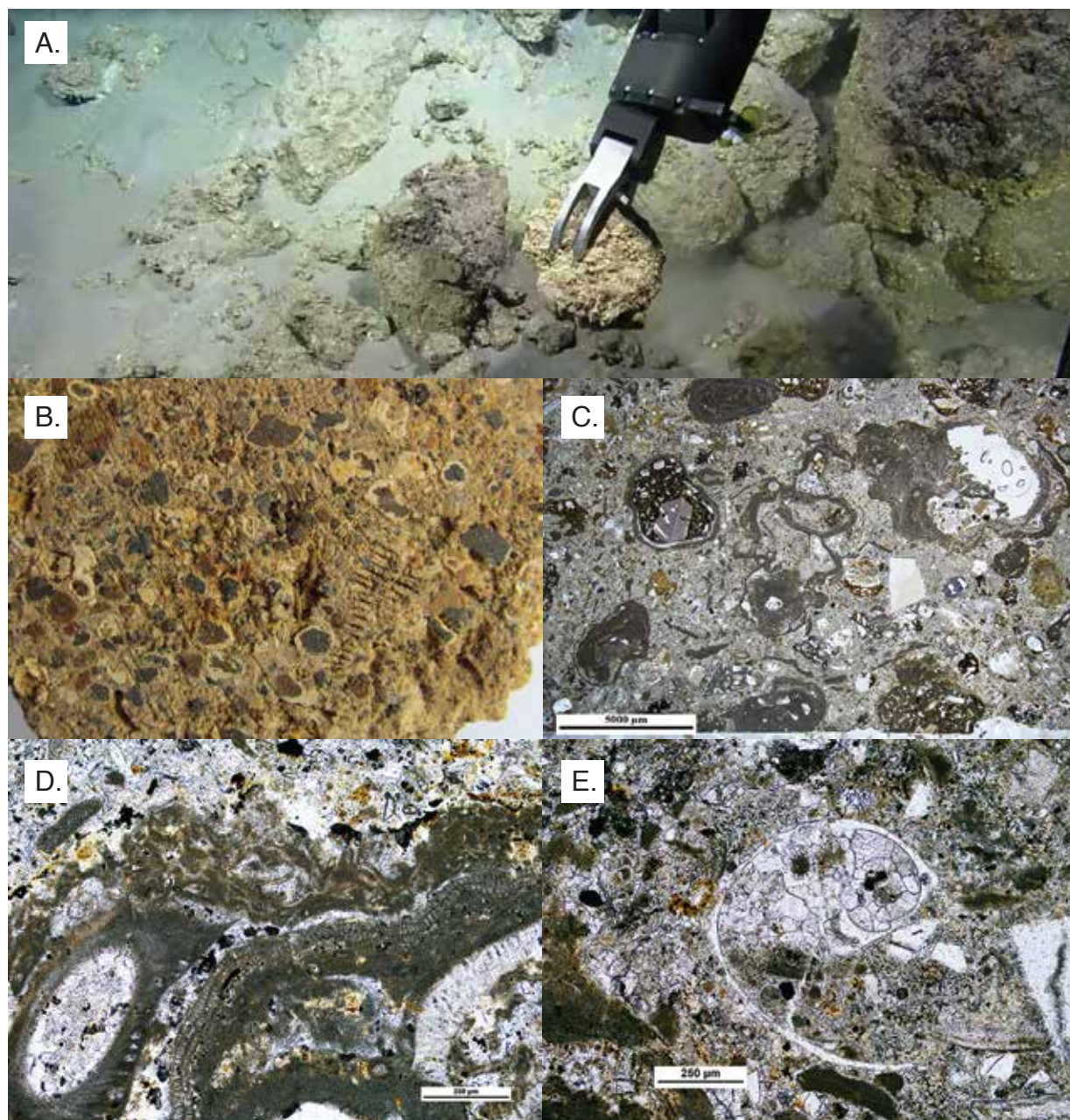


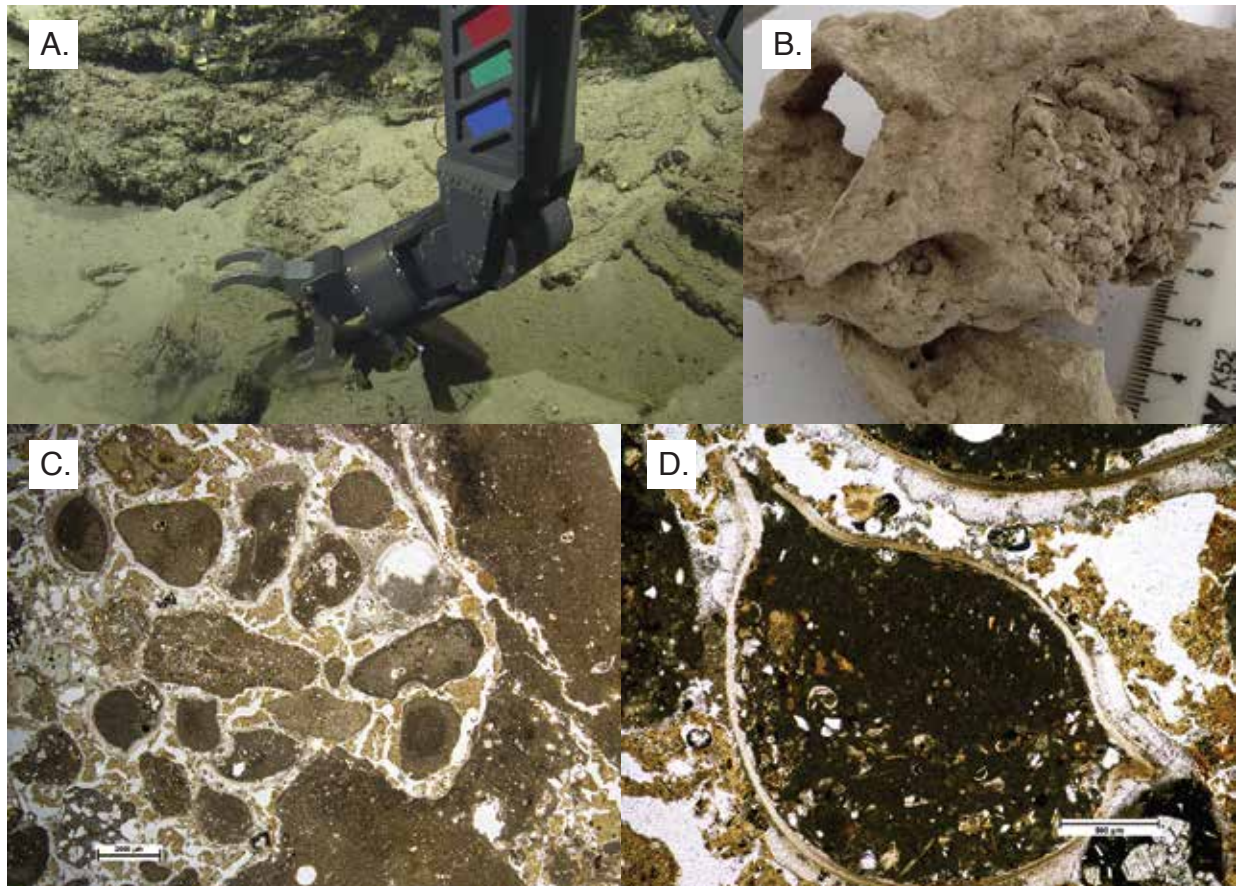


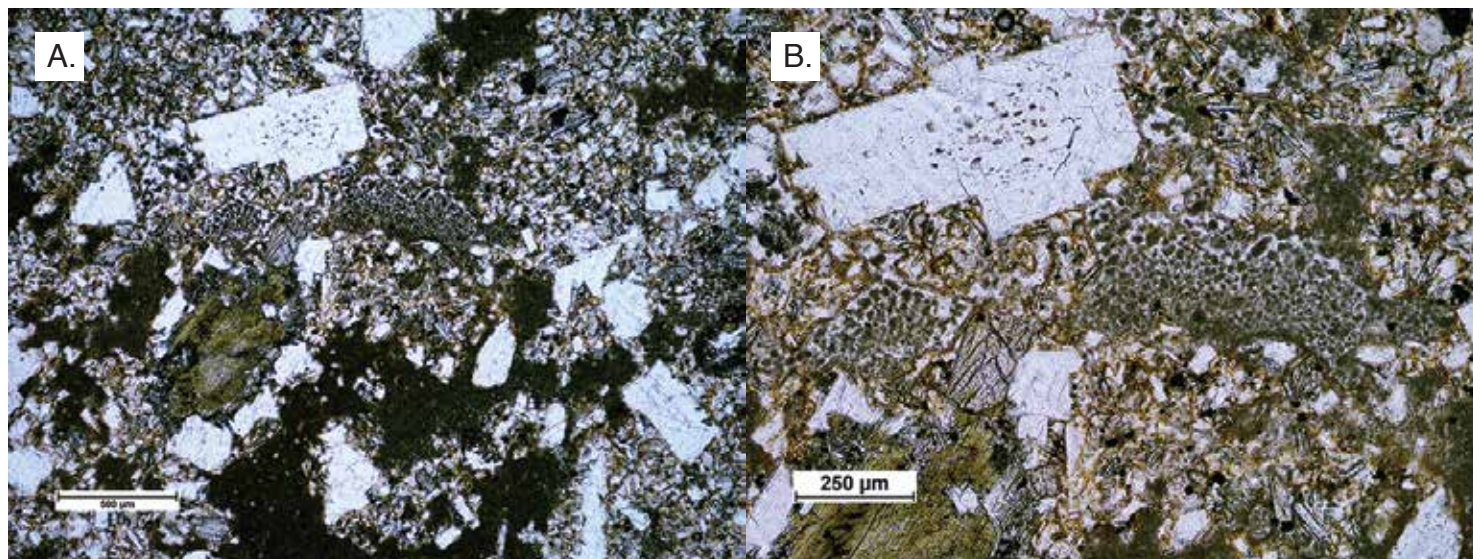


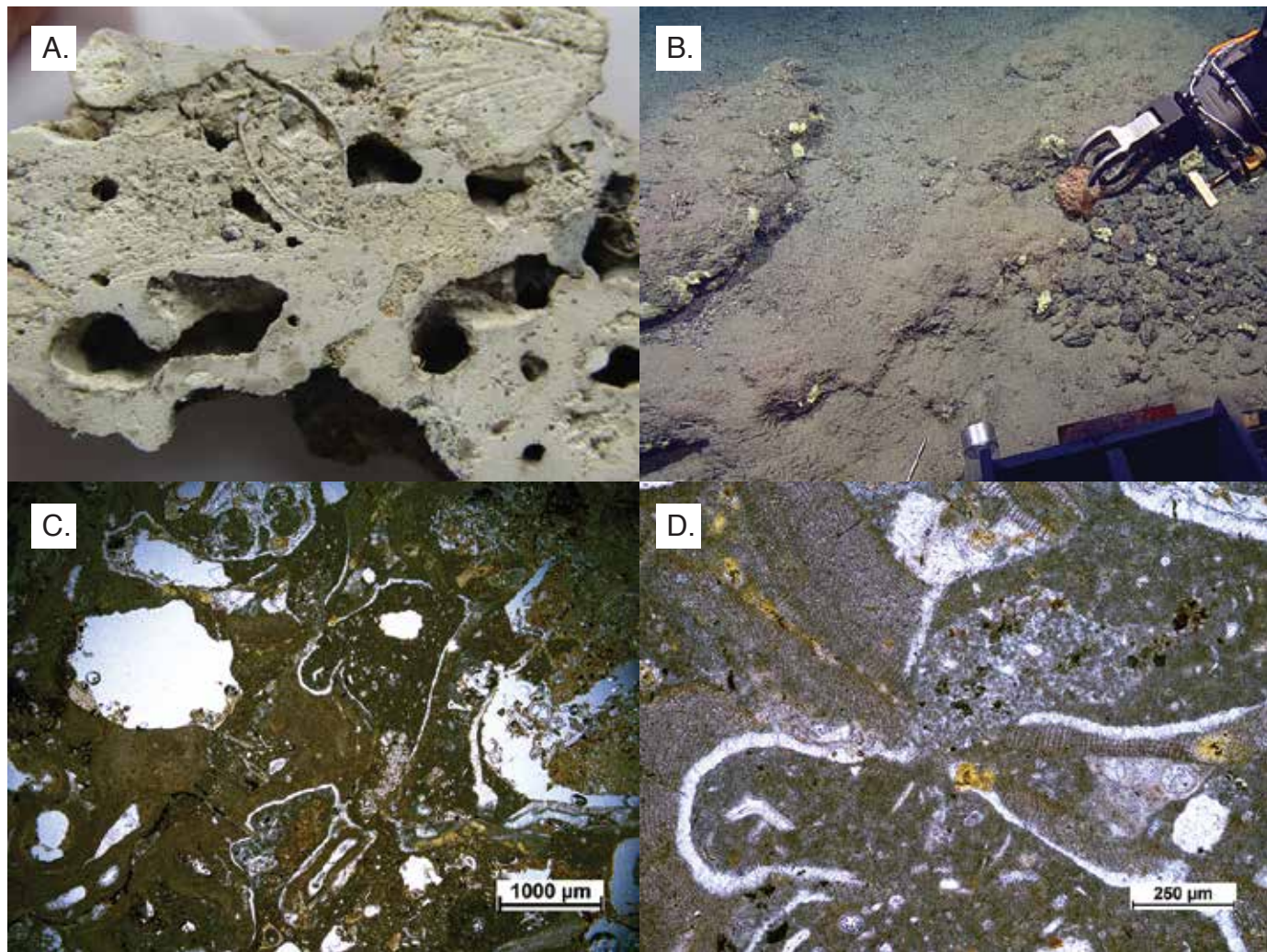


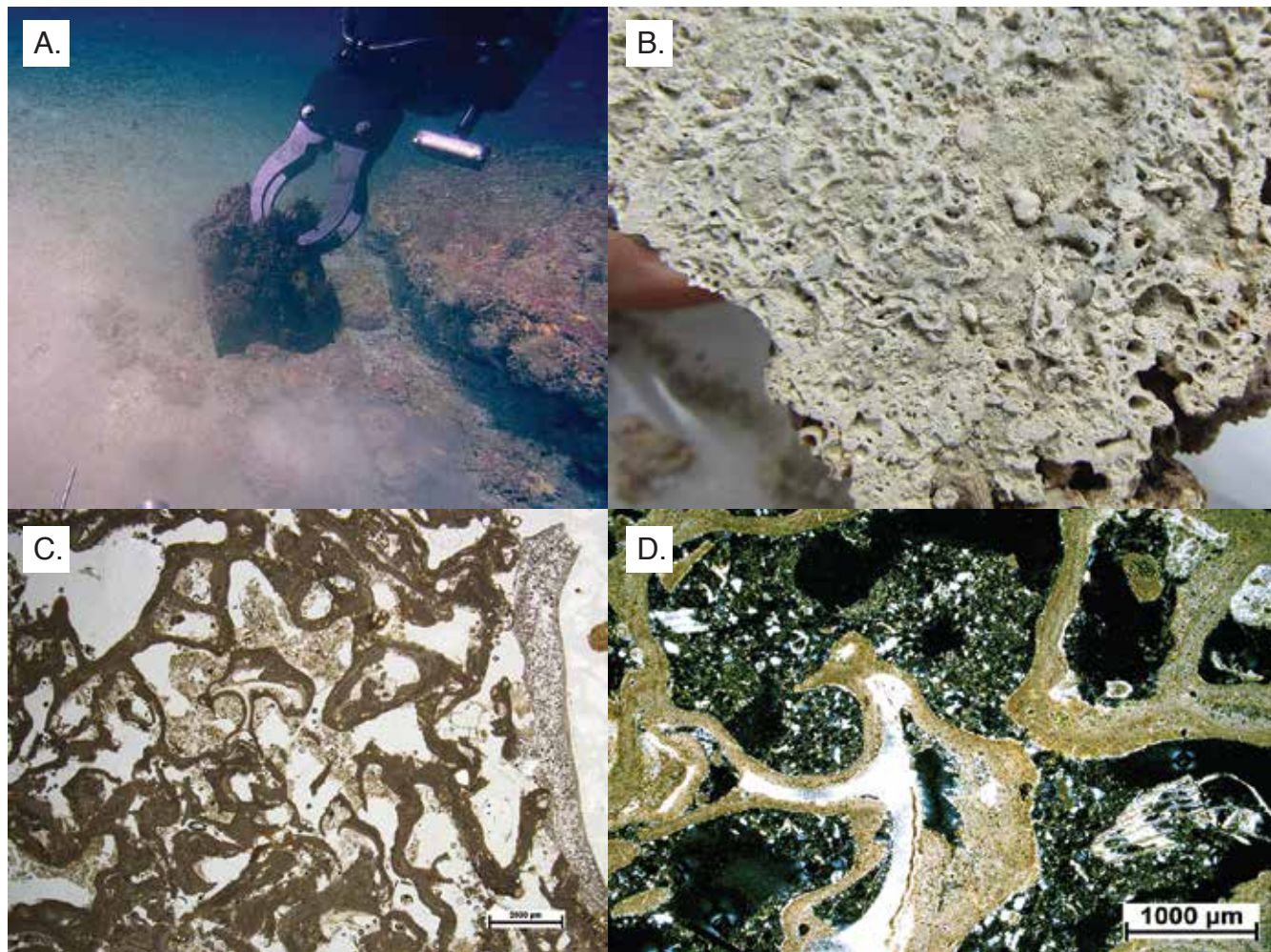












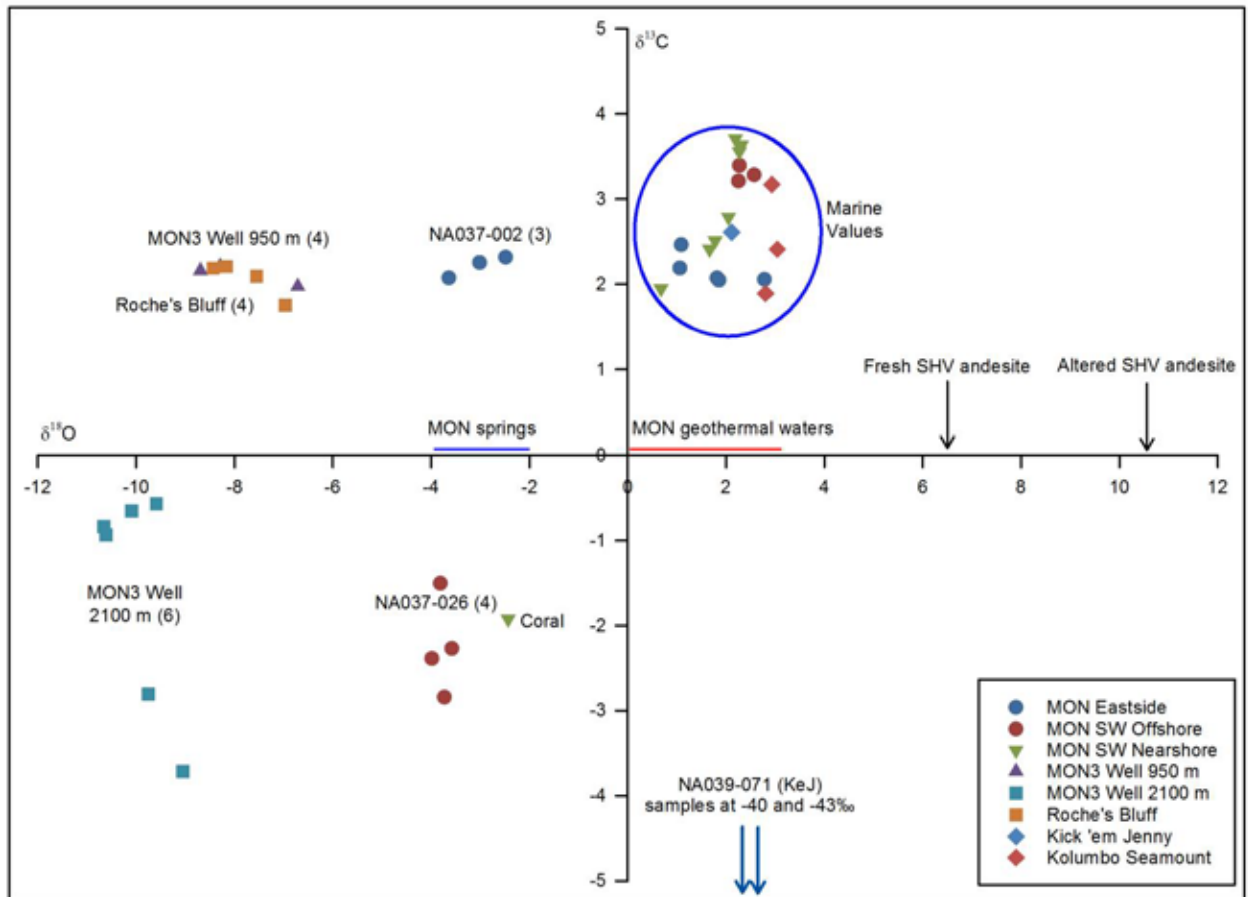
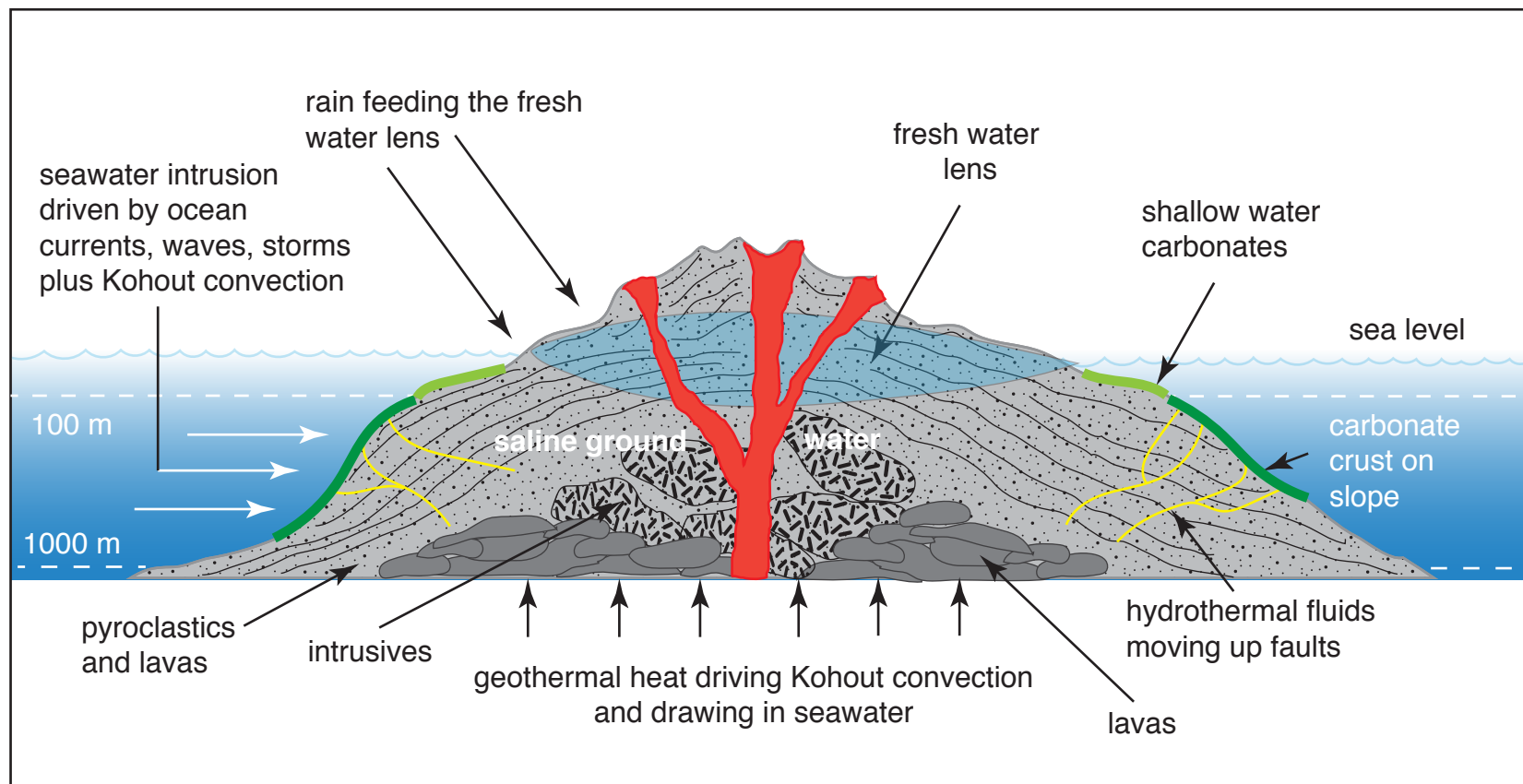


Figure 13

Fig. 13





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Conflicts of Interests

None of the authors has any financial or personal relationships with other people or organizations that could inappropriately influence their work.