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Abstract: An 8 m high wall in a quarry on the west end of Cayman Brac exposes the upper part of the Cayman Formation (Miocene), the lower part of the overlying Pedro Castle Formation (Pliocene), and the Cayman Unconformity, which is a karstic unconformity that separates these formations. The modern-day karst surface caps the Pedro Castle Formation. This exposure also includes cross-sections through two filled caves - the "Lower Cave" (> 8 m long, up to 2.5 m high) and "Upper Cave" (>23 m long, up to 2 m high) that are housed in the Cayman Formation and Pedro Castle Formation, respectively.

The Lower Cave is filled with caymanite, which is formed of laminated, varicoloured dolomitized mudstones and grainstones that contain scattered marine fossils (e.g., foraminifera, red algae). This cave, connected to the Cayman Unconformity by a small-diameter tunnel, evolved as part of the karst system that developed during the Messinian lowstand. The cave was filled and dolomitized prior to deposition of the Pedro Castle Formation. The Upper Cave is filled with a wide spectrum of lithotypes, including dolostones, calcareous mudstones, terra rossa, gastropod coquina, coated grains, and speleothems. U/Th dating indicates that some of the flowstones are >500,000 years old whereas others are only ~ 21,000 years old. Dolostones and mudstones in the basal part of the Upper Cave contain marine fossils (foraminifera, red algae) whereas the younger deposits are devoid of such fossils.

The Upper Cave and its deposits developed after the sediments of the Pedro Castle Formation had been deposited and lithified. Development of the cave filling deposits, which includes a clear transition from marine to non-marine influences, was controlled by eustatic sea-level changes and/or westward tectonic tilting of Cayman Brac that occurred after the Pedro Castle Formation became exposed, probably during the Late Pliocene.

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**CAVE-FILLS IN MIOCENE–PLIOCENE STRATA ON CAYMAN BRAC, BRITISH  
WEST INDIES: IMPLICATIONS FOR THE GEOLOGICAL EVOLUTION OF AN  
ISOLATED OCEANIC ISLAND**

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Key Words: Cave, cave deposits, terra rossa, flowstone, karst

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**31 ABSTRACT**

32 An 8 m high wall in a quarry on the west end of Cayman Brac exposes the upper part of the  
33 Cayman Formation (Miocene), the lower part of the overlying Pedro Castle Formation  
34 (Pliocene), and the Cayman Unconformity, which a karstic unconformity that separates these  
35 formations. The modern-day karst surface caps the Pedro Castle Formation. This exposure also  
36 includes cross-sections through two filled caves – the “Lower Cave” (> 8 m long, up to 2.5 m  
37 high) and “Upper Cave” (>23 m long, up to 2 m high) that are housed in the Cayman Formation  
38 and Pedro Castle Formation, respectively.

39 The Lower Cave is filled with caymanite, which is formed of laminated, varicoloured  
40 dolomitized mudstones and grainstones that contain scattered marine fossils (e.g., foraminifera,  
41 red algae). This cave, connected to the Cayman Unconformity by a small-diameter tunnel,  
42 evolved as part of the karst system that developed during the Messinian lowstand. The cave was  
43 filled and dolomitized prior to deposition of the Pedro Castle Formation. The Upper Cave is  
44 filled with a wide spectrum of lithotypes, including dolostones, calcareous mudstones, terra  
45 rossa, gastropod coquina, coated grains, and speleothems. U/Th dating indicates that some of the  
46 flowstones are >500,000 years old whereas others are only ~ 21,000 years old. Dolostones and  
47 mudstones in the basal part of the Upper Cave contain marine fossils (foraminifera, red algae)  
48 whereas the younger deposits are devoid of such fossils.

49 The Upper Cave and its deposits developed after the sediments of the Pedro Castle  
50 Formation had been deposited and lithified. Development of the cave filling deposits, which  
51 includes a clear transition from marine to non-marine influences, was controlled by eustatic sea-  
52 level changes and/or westward tectonic tilting of Cayman Brac that occurred after the Pedro  
53 Castle Formation became exposed, probably during the Late Pliocene.

## 54 1. Introduction

55 Isolated oceanic islands, like the Cayman Islands, are commonly formed of unconformity-  
56 bounded limestone/dolostone sequences that formed in response to oscillating sea levels (e.g.,  
57 Esteban and Klappa, 1983; Mylroie and Carew, 1995; Choquette and James, 1998; Liang and  
58 Jones, 2014). Although the unconformities in these successions may represent long periods of  
59 time it is commonly difficult to obtain information about the processes that were operative  
60 during those periods, largely because erosion and substrate modification predominate (e.g., Clari  
61 et al., 1995; Hillgartner, 1998; Sattler et al., 2005; Alonso-Zarza and Wright, 2010; Liang and  
62 Jones, 2015b). Joints, sinkholes, caves, and cavities, which are integral components of karst  
63 terrains, become receptacles where sediments can be deposited and speleothems precipitated  
64 (Jones and Smith, 1988; Jones, 1992b). Such deposits " . . . are of great importance, because  
65 once formed they are more protected from subsequent erosion than the contemporaneous surface  
66 sediments. They may thus preserve sediments and faunal remains for periods from which all  
67 other records are lacking" (Smart et al., 1988, p. 159). Sediments that accumulate in caves may  
68 come from multiple sources (e.g., Fornós et al., 2009, their Fig. 13; Farrant and Smart, 2011; van  
69 Hengstum and Scott, 2012; Fornós et al., 2014, their Fig. 10) and therefore have the potential of  
70 yielding information about the surface processes that were ongoing while the islands were  
71 exposed to the atmosphere.

72 This study focuses on a Miocene–Pliocene succession that is well exposed in the West End  
73 Quarry on Cayman Brac (Fig. 1). There, the quarry walls (up to 8 m high) include excellent  
74 exposures of the upper part of the Cayman Formation (Miocene), the Pedro Castle Formation  
75 (Pliocene), and two unconformities. The Cayman Unconformity is the boundary between the  
76 two formations (Jones and Hunter, 1994b) whereas the top of the quarry wall is the modern karst  
77 surface that caps the Pedro Castle Formation (Fig. 2). The east wall of the quarry includes

78 exposure of (1) a filled cave in the Cayman Formation that developed in association with the  
79 Cayman Unconformity and predated deposition of the Pedro Castle Formation, (2) a filled cave  
80 in the Pedro Castle Formation that postdated deposition of that formation, and (3) filled cavities  
81 that crosscut the Pedro Castle Formation and the Cayman Formation and parts of the filled caves.

82 Although numerous studies of cave-filling deposits have been presented (e.g., Ginés et al.,  
83 1981; Goodfriend and Mitterer, 1993; Foos et al., 2000; Cowie and Grant-Mackie, 2004; Ginés  
84 and Ginés, 2007; Auler et al., 2009; Fornós et al., 2009; Farrant and Smart, 2011; Martini, 2011;  
85 van Hengstum et al., 2011; Iacoviello and Martini, 2012; van Hengstum and Scott, 2012;  
86 Iacoviello and Martini, 2013; Fornós et al., 2014), little attempt has been made to integrate that  
87 information with the development of the bedrock succession that house the caves. Accordingly,  
88 this study, using the succession on Cayman Brac as an example, demonstrates the advantages of  
89 integrating the paragenetic histories of the bedrock succession and the cave-filling deposits for  
90 determining the geological evolution of an isolated island. In particular, such an analysis  
91 provides information on the processes that were operative during sea level lowstands when the  
92 island was exposed and karst and soils were developing under subaerial conditions.

## 93 **2. Geological setting**

94 Cayman Brac is a small, isolated island surrounded by deep waters of the Caribbean Sea  
95 (Fig. 1A). The elevated core of the island, which slopes westward from ~ 40 m above sea level  
96 (asl) at its east end to sea level at its west end, is skirted by a low-lying platform that is 2-3 m asl  
97 (Fig. 1B, C). The core of the island is formed of limestones and dolostones that belong to the  
98 Bluff Group (Fig. 2) whereas the peripheral platform is formed of limestones that belong to the  
99 Ironshore Formation (Fig. 1B).

100 This study is based on an exposure in the West End Quarry (SQA), which is an actively  
101 producing quarry located on the west end of Cayman Brac (Fig. 1B). The quarry walls, 6 to 8 m  
102 high, provide excellent sections through the upper part of the Cayman Formation (Miocene), the  
103 lower part of the Pedro Castle Formation (Pliocene), and the Cayman Unconformity (Fig. 3).  
104 The pervasively dolomitized Cayman Formation is characterized by skeletal  
105 wackestones/packstones that contain numerous massive (e.g., *Porites*, *Diploria*, *Montastrea*,  
106 *Favia*) and branching (e.g., *Stylophora*, *Porites*) corals along with red algae, bivalves, and  
107 gastropods (MacNeil and Jones, 2003). The Pedro Castle Formation is formed largely of skeletal  
108 wackestones that are characterized by foraminifera, red algae, branching corals (*Stylophora*,  
109 *Porites*) and scattered massive (*Montastrea*) corals (MacNeil and Jones, 2003). On this part of  
110 the island, the basal 0.5 to 1.0 m of the Pedro Castle Formation is formed of dolostone that is  
111 overlain by dolomitic limestone that is, in turn, overlain by limestone (Jones, 1994; Jones and  
112 Hunter, 1994a; MacNeil and Jones, 2003). Two unconformities are evident in this quarry wall.  
113 The Cayman Unconformity developed during the Messinian lowstand (Jones and Hunter, 1994b;  
114 Liang and Jones, 2015a), whereas the other unconformity, 3 to 4 m above the Cayman  
115 Unconformity, is the present day karst surface that is characterized by a rugged karst landscape  
116 (Liang and Jones, 2014).

### 117 3. Methods

118 The stratigraphic succession, location of the Cayman Unconformity, and the caves were  
119 mapped in the field. For ease of reference the filled caves in the Cayman Formation and Pedro  
120 Castle Formation are herein called the “Lower Cave” and “Upper Cave” respectively.

121 Orientated samples were collected from the bedrock and all of the cave-filling deposits.  
122 The basic petrography of the rocks was established from 16 large (7 x 5) and 7 small thin

123 sections (4.5 x 2.5 cm) made from samples impregnated with blue epoxy. Thin sections were  
124 stained with Alizarin Red S solution in order to separate the calcite from the dolomite.

125 The mineralogy of each sample was confirmed by X-ray diffraction analysis. Powdered  
126 samples, each weighing ~ 1 g, were analyzed on a Rigaku Ultima IV Powder X-ray system that  
127 was run at 38 kV and 38 mA using an Ultima IV X-ray generator with a Co tube. All scans were  
128 run from 5 to 90° 2 $\theta$  at a speed of 2°  $\theta$ /min. Mineral identifications were derived using the  
129 JADE 9.5 computer program that operates in tandem with the X-ray system.

130 Selected samples were examined on a Zeiss Sigma 300 VP-FESEM scanning electron  
131 microscope (SEM) that can be operated under high vacuum and variable pressure. Samples  
132 examined on the SEM included (1) fractured samples, up to 3 cm<sup>3</sup>, that were broken off the  
133 parent sample, (2) small and large uncovered thin sections, (3) rock slices, up to 4 x 4 x 0.5 cm,  
134 cut from a sample and polished on one surface, or (4) small samples (~ 1 cm<sup>2</sup>) with micro-milled  
135 surfaces that were prepared using a Fischione SEM Mill Model 1060 using 4kV and 50% focus  
136 with the sources positioned to four degrees and the sample continuously rotated.

137 Some samples examined on the SEM were left uncoated whereas others were coated with  
138 carbon. Images were obtained under variable conditions that depended on the sample. Some  
139 images are in normal mode whereas others are backscattered electron (BSE) images. Spot  
140 analyses, line transects, and elemental maps were obtained using a high resolution Bruker dual  
141 detector for energy-dispersive X-ray spectroscopy (EDS) system that is attached to the SEM.  
142 These standard-less, semi-quantitative analyses must be treated with caution given that the  
143 weight-percent values are probably  $\pm 5\%$ . Accordingly, these data were used largely to delineate  
144 trends and major changes in elemental contents that are apparent along transect lines or from the  
145 comparison between different points.

146  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios were obtained for four samples of dolostone in the Radiogenic Isotope  
147 Laboratory at the University of Alberta using the procedures described by MacNeil and Jones  
148 (2003). These analyses supplemented the analyses reported in MacNeil and Jones (2003) and  
149 Zhao and Jones (2012). All data were normalized to SRM 987 (0.710245). The error margin for  
150 these analyses is  $\pm 0.00002$ .

151 Twenty  $\text{O}^{18}$  and  $\text{C}^{13}$  isotope analyses, done by Isotope Tracer Technologies Inc. (Waterloo,  
152 Canada), were obtained using a DELTA<sup>Plus</sup> XL Stable Isotope Ratio Mass Spectrometer (IRMS)  
153 that is coupled with a ConFlo III interface and EA1110 Elemental Analyzer. All results are  
154 reported against the Vienna Pee Dee Belemnite (VPDB). Standards were run before, during, and  
155 after analysis of the samples in order to maintain accuracy. The error margin for the  $\text{O}^{18}$  and  $\text{C}^{13}$   
156 is  $\pm 0.1\%$ . These analyses supplement the analyses reported by MacNeil and Jones (2003) and  
157 Zhao and Jones (2012).

158 Three samples of flowstone were subject to U/Th dating. These analyses were analyzed by  
159 Dr. Bassam Ghaleb induction-coupled mass spectrometry (ICPMS) at GEOTOP, Montreal,  
160 Quebec. These samples had low U concentrations and traces of  $^{232}\text{Th}$ , which indicates that  
161 detrital material had been incorporated into the precipitated calcite. Thus, the calculated ages  
162 were corrected for this using the methodology of Ludwig and Paces (2002).

### 163 **3. Terminology**

164 Caymanite, named after local artisans who use it to make jewellery, is a multicoloured  
165 (white, red, black) finely crystalline dolostone with laminae that dip at angles up to  $60^\circ$  (Jones,  
166 1992a). This cavity-filling internal sediment is common in the Cayman Formation. Although  
167 mentioned by Folk and McBride (1976), Rigby and Roberts (1976), and Lockhart (1986), it was  
168 examined in detail by Jones (1992a).

169 The term limpid dolomite, following Folk and Siedlecka (1974), Folk and Land (1975),  
170 Longman and Mench (1978), and Kaldi and Gidman (1982), is applied to water-clear dolomite  
171 crystals that commonly formed as a cement.

172 The term “terra rossa”, first used by Tućan (1912) for the distinctive red soils found in the  
173 Mediterranean region. Merino and Banerjee (2008, p. 62) stated that terra rossa, known from  
174 many areas throughout the world, are “... red claystones up to several metres thick and  
175 kilometers across that occur at the earth’s surface and are associated with karst carbonates”. In  
176 some areas, including Jamaica and France, terra rossa grades into bauxite (Merino and Banerjee,  
177 2008). According to Durn et al. (1999), terra rossa is typically formed of quartz, plagioclase, K-  
178 feldspar, micaceous clay minerals, kaolinites, chlorite, vermiculite, smectite, hematite, goethite,  
179 XRD-amorphous inorganic compounds, and in some cases, calcite, dolomite, and boehmite.  
180 Bauxite is typically formed of gibbsite ( $(\text{Al}(\text{OH})_3)$ ), amorphous Al hydroxides, boehmite ( $\gamma$   
181  $\text{AlO}(\text{OH})$ ), diaspose ( $\alpha$   $\text{AlO}(\text{OH})$ ), hematite ( $\text{Fe}_2\text{O}_3$ ), goethite ( $\text{FeO}(\text{OH})$ ), anatase ( $\text{TiO}_2$ ), and  
182 kaolinite ( $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ ) (Boni et al., 2013).

183 Dolostones from the bedrock and the caves are divided into low-Ca calcium dolomite  
184 (LCD) that contains < 55 mol %  $\text{CaCO}_3$  and high-Ca calcium dolomite (HCD) that contains >55  
185 mol %  $\text{CaCO}_3$  (Jones et al., 2001). Following the convention adopted by Jones et al. (2001), the  
186 mol %  $\text{CaCO}_3$  is abbreviated to %Ca.

## 187 **4. Results**

### 188 *4.1. Cave morphologies*

189 The north-south oriented wall on the east side of West End Quarry has its base at 1.5 m, the  
190 Cayman Unconformity at 4.5 to 5.0 m, and its top at 7.5 m above sea level (asl). This quarry

191 wall includes sections through the Lower Cave and the Upper Cave (Fig. 4). Late stage cavities  
192 cross-cut both caves and the bedrock.

193 The Lower Cave, which has an irregular cross-sectional shape, is at least 8 m long (south  
194 end is covered) with a maximum height of 2.5 m (Fig. 4A). At its north end, this cave appears to  
195 be connected to the Cayman Unconformity by a small-diameter (< 0.5 m) tunnel, but poor  
196 outcrop conditions in that area precluded a definitive assessment.

197 The Upper Cave, with an irregular cross-sectional morphology, is at least 23 m long and up  
198 to 2 m high (Fig. 4B). The north end of the cave is poorly exposed and impossible to map in  
199 detail. This cave may be formed of two or three smaller chambers that are linked together by  
200 short tunnels, but poor outcrop conditions in this critical area prevented resolution of this issue.  
201 Although the base of the cave is typically coincident with or just above the Cayman  
202 Unconformity, it locally cuts down in the upper part of the Cayman Formation (Fig. 4B).

203 Small-diameter (< 2 m) subvertical sinkholes, located at the south end, the middle part, and  
204 possibly the north end of the exposure, extend down from the present-day surface (Fig. 4). The  
205 “Middle Sinkhole” intersects the Upper Cave (Fig. 4B), whereas the “South sinkhole” intersects  
206 the Upper Cave with parts extending down into the Lower Cave (Fig. 4A).

#### 207 4.2. *Lithotypes*

208 The dominant lithotypes in the caves, based on field appearance and thin section analyses,  
209 are caymanite, finely crystalline dolostone, varicoloured calcareous mudstones, red terra rossa,  
210 red terra rossa breccia, terra rossa ooids, olive terra rossa, gastropod coquina, coated grains, and  
211 speleothems (Figs. 5-7).

#### 212 4.2.1. *Caymanite*

213 Caymanite, restricted to the Lower Cave and small cavities in the dolostones of the  
214 Cayman Formation, is formed largely of white laminae and fewer, thin light to dark red and light  
215 grey laminae that dip at angles up to 60° (Fig. 4, 5). The caymanite is formed of very hard,  
216 dense HCD (56.7–57.3%Ca) that is composed of interlocking euhedral to subhedral crystals < 5  
217  $\mu\text{m}$  long (Figs. 8, 9). Although dominated by mudstone laminae, there are rare (< 1 cm thick)  
218 grainstone laminae that include fragments of red algae, small benthic foraminifera, planktonic  
219 foraminifera, and other unknown biofragments (Fig. 8). Some grainstone laminae include  
220 angular grains (< 1 mm long) of finely crystalline dolostone that were probably derived from the  
221 Cayman Formation (Fig. 8B). Some caymanite, especially in the upper part of the cave, contain  
222 pores (< 1 mm) lined with limpid dolomite cement and occluded by calcite cement (Fig. 9B).

#### 223 4.2.2. *Finely crystalline dolostone*

224 Units A and B that form the basal part of the succession in the Upper Cave are finely  
225 crystalline dolostones (Fig. 6A). Unit A (20 cm thick) is white with vaguely defined laminae in  
226 the upper part, whereas unit B is characterized by off-white to very light pink horizontal laminae  
227 (Fig. 6A). They differ from caymanite because they (1) lack the multicolored laminae that dip at  
228 various angles, and (2) are “softer” and lack the porcellaneous appearance of caymanite.

229 The dolostone in Unit A, formed of subhedral to euhedral HCD (56.1 %Ca) crystals < 2  $\mu\text{m}$   
230 long (Fig. 10A-C), contains scattered dolomitized fragments of red algae (Fig. 10C). Small  
231 pores in the dolostone are lined with limpid dolomite cement and occluded by calcite (Fig. 10A).

232 The dolostone in unit B is formed of euhedral HCD (57.8 %Ca) crystals < 5  $\mu\text{m}$  long (Fig.  
233 10D-F). It contains scattered dolomitized red algae fragments and pores lined with limpid

234 dolomite cement and filled with calcite. Unlike unit A, hollow dolomite crystals are common in  
235 the upper part of this unit (Fig. 10F).

#### 236 4.2.3. *Calcareous mudstone*

237 The white to pink to red calcareous mudstones (Unit C, Fig. 6A, B, C), which overlie the  
238 dolostones of units A and B in the Upper Cave, are formed of anhedral to subhedral calcite  
239 crystals that are  $< 2 \mu\text{m}$  long with most being  $< 1 \mu\text{m}$  long (Fig. 11, 12A-C). No dolomite was  
240 detected in these mudstones. Some of the mudstones contain scattered foraminifera that are up  
241 to 0.5 mm long (Fig. 11C, D). The mudstones, typically forming beds  $< 2$  cm thick, are  
242 structureless or finely laminated with graded bedding (Fig. 11B). Some of the mudstone beds  
243 show evidence of desiccation, brecciation, and clast rotation (Fig. 11E, F).

244 Pores and irregular fractures in the mudstones are lined or filled with calcite cement (Fig.  
245 11A). No limpid dolomite cement was found. Larger cavities commonly have small lithoclasts  
246 on their floors that were then buried by mudstone, which were, in turn, coated by calcite cement  
247 (Fig. 11F). Many of the larger cavities were not completely filled.

248 All of the mudstones, which range from white to off-white to pink to red (Fig. 11A, B, C),  
249 contain scattered Fe-rich, ovate to spherical masses, that are up to  $20 \mu\text{m}$  (typically  $< 10 \mu\text{m}$ ) in  
250 diameter (Fig. 12D-L). The red mudstones have the highest concentration of Fe-masses whereas  
251 the white mudstones have the lowest. The microstructure of these Fe-rich masses ranges from  
252 triangular-shaped “crystals” (Fig. 11G) to “plates” (Fig. 11F) to structureless masses (Fig. 11I).  
253 EDS analyses show that they contain Fe (62-67%), O (27-30%), Al (2-4%), Si (1-2%), and Ca  
254 (1-3%) whereas the platy crystals contain 47% Fe (Fig. 13). The Ca is probably derived from  
255 minute particles of the matrix that are embedded in the Fe-rich precipitates.

256 4.2.4. *Red terra rossa*

257 The colour contrast between the red lithified terra rossa and the white dolostones of the  
258 bedrock makes it the most obvious deposit in the caves (Figs. 4, 5D, 6B, E). XRD analyses  
259 shows that it is formed of calcite, boehmite, gibbsite, anatase, and minor amounts of kaolinite.  
260 The calcite, however, is restricted to cobweb-like arrays of microfractures, typically < 0.1 mm  
261 wide, that radiate throughout the terra rossa (Fig. 14). Element maps emphasize the spatial  
262 discrimination with the Al, Si, Fe and other allied elements being restricted to irregularly shaped  
263 masses, typically < 0.5 mm long, that are surrounded by the calcite-filled fractures (Fig. 14D-F,  
264 15). There is commonly more than one generation of calcite cement (Fig. 14G-I). In these  
265 cases, the larger masses of terra rossa are cut by microfractures that are filled with fibrous calcite  
266 that in plane polarized light is off-white to yellowish in colour (Fig. 14G, I). In contrast, the  
267 wider fractures around the larger masses are filled with calcite that is colourless in plane  
268 polarized light (Fig. 14G, I). Textural evidence indicates that this calcite formed after the off-  
269 white to yellowish calcite. EDS transects clearly highlight the compositional contrasts between  
270 the red terra rossa and the calcite (Figs. 14H, 15).

271 Red terra rossa imaged by standard SEM analyses of fractured and polished surfaces,  
272 appears amorphous and EDS spot and line analyses show that it is formed largely of Al, Fe, and  
273 Si (Fig. 15). Imaging and backscatter electron imaging of highly polished micro-milled samples,  
274 however, showed that the terra rossa has the following characteristics.

- 275 • The terra rossa is formed largely of lath- to plate-shaped crystals < 2  $\mu\text{m}$  long and 250 nm  
276 wide but mostly < 250 nm long and < 100 nm wide (Figs. 16A-C). EDS analyses show  
277 that they are formed of Al and Si but do not contain Fe. Although compositionally

278 consistent with boehmite and gibbsite (as identified by XRD analysis) it proved impossible  
279 to precisely identify which crystals are which minerals.

- 280 • The Fe is located in Fe-rich masses, < 5  $\mu\text{m}$  in diameter (rare), and irregular-shaped grains,  
281 < 1  $\mu\text{m}$  long and commonly < 100 nm long, that are randomly disseminated throughout the  
282 rock. These grains are assumed to be Fe-oxides because EDS failed to detect S in any of  
283 them. There is no obvious pattern to the distribution of the Fe.
- 284 • Throughout the terra rossa there are scattered exotic crystals/grains that are formed of (a)  
285 zirconium silicate, < 12  $\mu\text{m}$  long (Fig. 16F), (b) Ti-oxides, < 15  $\mu\text{m}$  long (Fig. 16G), (c) Cr  
286 and Fe (Fig. 16H) grains < 25  $\mu\text{m}$  long, and (d) P, Ce, and La grains < 5  $\mu\text{m}$  long (Fig.  
287 16I). There is no obvious distribution pattern to these grains.

#### 288 4.2.5. *Terra rossa breccia*

289 This distinctive rock, common in the upper part of the Upper Cave near the Middle  
290 Sinkhole, is formed of (1) irregular-shaped masses of red terra rossa up to 15 cm long and 10 cm  
291 high, (2) thinly laminated flowstone (up to 1 cm thick) that coats the terra rossa masses (Fig. 7C),  
292 and (3) calcareous mudstone that filled the areas between the flowstone (Fig. 7C). Irregular-  
293 shaped cavities, up to 5 cm high and wide, occur where the spaces between the terra rossa  
294 masses was not completely filled with flowstone and/or limestone (Figs. 7A, B).

295 Thin sections viewed in plane polarized light show terra rossa that is dark red to orange to  
296 yellowish brown in colour and dissected by irregular networks of calcite-filled fractures (Fig.  
297 17A). BSE images and EDS analyses, however, failed to show any obvious compositional  
298 differences between the different colours of terra rossa (Fig. 17B, C, 18). Likewise, there was no  
299 obvious difference in the amount and distribution of disseminated Fe in these areas (Fig. 17D,  
300 E).

#### 301 4.2.6. *Terra rossa ooids*

302 This lithotype comprises spherical to ellipsoidal ooids, up to 1 mm long, formed of red  
303 terra rossa held in spar calcite cement (Fig. 21). Although superficially uniform, these ooids are  
304 characterized by variable colours (as seen in plane polarized light), internal structures, and  
305 compositions (Figs. 21, 22). With plane polarized light, the ooids range from dark red to light  
306 reddish-orange (Fig. 21). Some are homogeneous with no apparent nucleus (Fig. 21A), whereas  
307 other ooids have a nucleus encased by a thin cortex (Fig. 21C) or a cortex that is irregular in  
308 thickness with multiple bands (Fig. 21E, G, I, K). Most nuclei are formed of terra rossa that  
309 appears denser and darker in colour relative to the surrounding cortex (Fig. 21E, J, L). In rare  
310 examples, small pieces of limestone form the nuclei (Fig. 21G).

311 BSE images and EDS analyses of the ooids show that, irrespective of colour, they are  
312 formed largely of Al, Si, and Fe with lesser amounts of Ca (Fig. 22). The lighter coloured  
313 cortical laminae, where present, reflect zones where Al, Si, Fe, and Ca mixed together in varying  
314 proportions (Fig. 22B, C, D, E, F). High magnification BSE images confirm that such zones are  
315 formed of interspersed calcite, clays, and Fe-oxide grains.

#### 316 4.2.7. *Olive terra rossa*

317 The olive terra rossa fills cavities that crosscut the caves and/or cavities that are filled with  
318 red terra rossa (Fig. 6E). The olive terra rossa is cut by honeycombed arrays of calcite-filled  
319 microfractures (Fig. 17D, E). Under plane polarized light this terra rossa ranges from light- to  
320 dark-yellow brown to bright red with no readily discernable pattern (Fig. 19A). The colours  
321 evident under plane polarized light are deceptive because there are no obvious correlations  
322 between the colours and elemental content as revealed by EDS analyses (Fig. 2). Some of the  
323 bright red areas, for example, have a high Ca but low Al-Fe-Si content (Figs. 19C, 20A) whereas

324 some of the brown-coloured areas have subequal amounts of Ca and Al-Fe-Si (Fig. 19E, 20B).  
325 Some of these “coloured” areas, like those shown in Figure 19B and 19C, are formed of small,  
326 irregular-shaped patches of clays and Fe that are scattered throughout a finely crystalline calcite  
327 groundmass. Thus, it is possible that the colour may, in some way, reflect the amount of calcite  
328 that is mixed in with the clays and Fe.

#### 329 4.2.8. *Gastropod coquina*

330 Found only in one small area in the uppermost part of the Upper Cave north of the Middle  
331 Sinkhole (Fig. 4B), this lithotype is characterized by numerous, closely packed gastropods that  
332 are up to 1.5 cm high and 1.5 cm in diameter (Fig. 7F). The gastropods are well preserved with  
333 no evidence of alteration of their aragonitic shells. In most cases, however, the shells have been  
334 dislodged (probably during quarrying) and impressions are all that remain (Fig. 7F). Although  
335 the species is unknown, this appears to be a monospecific assemble of gastropods.

#### 336 4.2.9. *Coated grains*

337 Coated grains, held in spar calcite cement, forms the (1) matrix between the gastropods in  
338 the gastropod coquina, and (2) bed that overlies the gastropod coquina. Up to 4 mm long, the  
339 ~~coasted~~ grains have various types of nuclei that are encased by cortical laminae formed of  
340 micrite that commonly contains small shell fragments (Fig. 23). Nuclei range from complete  
341 gastropod shells (Fig. 23A, B) to fragments of gastropod shells (Fig. 23C-E), to small limestone  
342 fragments (Fig. 23F). Some body cavities in the gastropods are partly filled with terra rossa  
343 ooids (< 1 mm diameter) and/or spar calcite cement (Fig. 23B). Vague laminations in the  
344 cortices are defined by subtle differences in colour (Fig. 23E, F). The oncooids are typically  
345 coated with a thin isopachous rind of calcite cement (~~Fig. 13 D~~). Pores between the oncooids are  
346 filled with terra rossa ooids, small shell fragments, and spar calcite cement (Fig. 23).

347 Despite careful examination of the oncoids at high magnifications on the SEM, no evidence  
348 of microbes was found in their ~~cortices~~,

#### 349 4.2.10. *Speleothems*

350 Speleothems in the Upper Cave include flowstone (Fig. 6E), stalactites and draperies (Fig.  
351 7D, E), and rare stalagmites. No speleothems were found in the Lower Cave.

352 In the lower part of the Upper Cave, brownish flowstone (up to 15 cm thick) is intercalated  
353 with the white to red mudstones (Fig. 6E) and, in some areas, thin beds of red terra rossa. The  
354 banded flowstone is formed largely of densely packed prismatic calcite crystals that are up to 2  
355 cm long and 5 mm wide. Exceptions to this motif include (1) laminae formed of small dendritic  
356 bushes, and (2) thin brown to dark grey micrite laminae that have a pseudo-stromatolitic  
357 appearance. No evidence of microbes was detected in these layers. There is no textural evidence  
358 to indicate that the calcite formed through alteration of aragonite.

359 The flowstone in the upper part of the cave, which coats the terra rossa, is thinly laminated  
360 and lacks the distinctive brownish colour of the lower flowstone (Fig. 7C).

361 Stalactites and draperies adorn the cave ceiling where the cave was not completely filled by  
362 sediments and flowstone (Fig. 7D, E). The stalactites and draperies, up to 15 cm long, are  
363 distinctive because of their surface white color (Fig. 7D, E). Sections through the stalactites and  
364 draperies show that they are thinly laminated and formed entirely of calcite.

365 Rare stalagmites, found only in the area below the Middle Sinkhole, are poorly developed  
366 and no more than 6 cm high and 5 cm in diameter.

367 4.3.  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios

368 Dolostones in the upper part of the Cayman Formation have  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of 0.70902 to  
369 0.70913 (Zhao and Jones, 2012) whereas the dolostones in the Pedro Castle Formation have  
370  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of 0.70903 to 0.70911 (MacNeil and Jones, 2003).

371 Additional  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios obtained during this study include values of (1)  $0.70905 \pm$   
372  $0.00003$  for dolostone from the Cayman Formation beside the Lower Cave, (2)  $0.70907 \pm$   
373  $0.00003$  for the caymanite in the Lower Cave, (3)  $0.70907 \pm 0.00002$  for the dolostone in Unit A  
374 of the Upper Cave, and (4)  $0.70913 \pm 0.00004$  for the dolostones in Unit B of the Upper Cave  
375 (Fig. 24).

376 4.4. Carbon and oxygen stable isotopes

377 According to Zhao and Jones (2012), dolostones in the upper part of the Cayman  
378 Formation on Cayman Brac have  $\delta^{13}\text{C}_{\text{VPDB}}$  values of 1.6 to 3.5‰ (average 2.5‰) and  $\delta^{18}\text{O}_{\text{VPDB}}$   
379 values of 2.3 to 4.0‰ (average 3.2‰), whereas MacNeil and Jones (2003) obtained  $\delta^{13}\text{C}_{\text{VPDB}}$   
380 values of -1.8 to +1.4‰ (average 0.3‰) and  $\delta^{18}\text{O}_{\text{VPDB}}$  values of -0.08 to +2.16‰ (average  
381 +1.3‰) from the dolostones from the Pedro Castle Formation. In this study, the isotope values  
382 obtained for dolostone from the Cayman Formation beside the Lower Cave yielded values within  
383 the range given by Zhao and Jones (2012). Values obtained from a dolomitic limestone from the  
384 Pedro Castle Formation bedrock beside the Upper Cave are more negative than those reported by  
385 MacNeil and Jones (2003) because it contained ~ 65% calcite (Fig. 24).

386 O and C isotopes values obtained from the cave deposits can be divided into groups A and  
387 B. Group A, which includes the dolostones from the Cayman Formation, the caymanite, and the  
388 dolostones in Units A and B in the Upper Cave, are characterized by similar values that are  
389 comparable to the values previously reported for the Cayman Formation (Fig. 24). The more

390 negative values for the caymanite in the upper part of the Lower Cave reflects the fact that it  
391 contains ~45% calcite cement. Similarly, the more negative values for the dolostones in the  
392 lower part of the Upper Cave probably reflects the fact that Units A and B contain ~ 5% and ~  
393 15% calcite cement, respectively.

394 Group B includes (1) all of the lithotypes that form the middle and upper parts of the  
395 succession in the Upper Cave, and (2) the red and olive terra rossa that fills cavities that crosscut  
396 deposits in the Lower Cave and Upper Cave and the bedrock of the Cayman Formation and  
397 Pedro Castle Formation (Fig. 24). For these deposits the  $\delta^{18}\text{O}$  ranges from -4.3 to -6.7‰ and the  
398  $\delta^{13}\text{C}$  ranges from -6.0 to -11.6‰ (Fig. 24).

#### 399 4.5. *U/Th dating*

400 Dating of two sample of flowstone (Fig. 6E) from the lower part of the Upper Cave  
401 indicated that it was more than 500,000 years old.

402 A speleothem sample from the uppermost part of the Upper Cave (near the Middle  
403 Sinkhole) is formed of three parts as defined by colour differences and separation by  
404 discontinuities. U/Th dating of each part yielded the following ages (1) >500,000 years for the  
405 basal flowstone layer, (2)  $61,953 \pm 1,577$  years for the basal, incipient stalagmite (4.0 cm in  
406 diameter, 0.6 cm high), and (3)  $22,303 \pm 2,310$  years and  ~~$19,261 \pm 1,938$~~  years for two samples  
407 taken from the uppermost part of the stalagmite (4.0 in diameter, 0.7 cm high).

#### 408 **5. Dolomitization**

409 According to Zhao and Jones (2012, their Fig. 12), two phases of dolomitization (I and II),  
410 both mediated by seawater, affected the limestones of the Cayman Formation and Pedro Castle  
411 Formation. Based on  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, petrographic evidence, and stratigraphic relationships they  
412 offered the following conclusions.

- 413 • Phase I occurred in the Late Miocene (6 to 8 Ma) and Phase II occurred during the Pliocene  
414 to Early Pleistocene (1 to 5 Ma, but with mode at 2 to 4.5 Ma).
- 415 • Phase I dolomitization resulted in partial dolomitization of the Cayman Formation.
- 416 • Phase II dolomitization completed dolomitization of the Cayman Formation and  
417 dolomitized the lower part of the Pedro Castle Formation.
- 418 • Phase II dolomitization took place while sediments of the Pedro Castle Formation were still  
419 accumulating.
- 420 • Phase II dolomitization was completed before Cayman Brac was tectonically tilted.

## 421 **6. Geological evolution of lower cave**

422 The notion that the Lower Cave was genetically linked to the Cayman Unconformity that  
423 developed during the Messinian lowstand (Jones, 1994; Jones and Hunter, 1994b; Liang and  
424 Jones, 2015a) is based on (1) the location of the cave just beneath the unconformity and, (2) at  
425 the north end of the cave, there appears to be a short, small-diameter (< 0.5 m) tunnel that  
426 connects the cave to the unconformity. The latter suggestion, however, must be treated with  
427 some caution because outcrop conditions in that critical area are poor. This architecture is  
428 parallel to the situation found in Pedro Castle Quarry on Grand Cayman where a filled cave in  
429 the upper part of the Cayman Formation was linked to the Cayman Unconformity (Jones and  
430 Smith, 1988, their Fig. 6; Jones, 1992b, his Fig. 2).

431 The Lower Cave is filled with caymanite, which is not found in the Upper Cave. Although  
432 common in the Cayman Formation, caymanite is rare in the Pedro Castle Formation (Jones,  
433 1992a). The original sediments, derived from nearby marine sediments, soils, and swamps were  
434 probably transported into caves during severe storms that generated large sea waves that

435 inundated low-lying parts of the island (Jones, 1992a) and/or by surface run-off following heavy  
436 rainfall. The presence of marine biofragments attests to the marine influence.

437         The dolostone that forms the caymanite has stable (O and C) and radiogenic ( $^{87}\text{Sr}/^{86}\text{Sr}$ )  
438 isotopes similar to those derived from the dolostones of the Cayman Formation (Fig. 24). The  
439  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios from the caymanite are consistent with Phase I dolomitization as defined by Zhao  
440 and Jones (2012), which took place 6 to 8 Ma (Late Miocene). These similarities indicate that  
441 (1) the original sediments in the cave were deposited before dolomitization took place, and (2)  
442 dolomitization of the bedrock and the cavity-filling sediments took place at the same time.

## 443 **7. Geological evolution of Upper Cave**

444         The Upper Cave is housed in the Pedro Castle Formation except for local areas where the  
445 cave floor was cut down into the Cayman Formation (Fig. 4B). The lateral and vertical  
446 variability in lithotype distribution in this cave means that it is difficult to produce a single  
447 stratigraphic succession that is applicable to all parts of the cave. Nevertheless, a composite  
448 section based on available information can be divided into packages I to V (Fig. 24) that reflect  
449 the distinct stages involved in the filling of the cave. All of these deposits must be younger than  
450 the cave that postdated deposition of the sediments that now form the Pedro Castle Formation.

### 451 *7.1. Package I (P-I)*

452         This package, 30 cm thick, includes the dolostones of units A and B that contain marine  
453 biofragments, including red algae and foraminifera. The marine sediments were transported  
454 when the cave was flooded by marine water or when storms washed marine sediment on land  
455 and into the cave.

456         The  $^{87}\text{Sr}/^{86}\text{Sr}$  of these dolostones are similar to those from the dolostones in the Pedro  
457 Castle Formation but must be younger because they are in a cave housed in that formation.

458 *7.2. Package II (P-II)*

459 This package is formed of varicoloured calcareous mudstones (Fig. 6E) that contain  
460 scattered foraminifera (Fig. 11C, D). Locally, this unit has a chaotic appearance because many  
461 of the mudstone beds were brecciated, probably as a result of desiccation and/or erosion as fast-  
462 flowing water moved through the cave. Spaces between the rotated clasts are commonly filled  
463 with varicoloured mudstones and various generations of calcite cement (Fig. 6B-D).

464 The boundary with the underlying dolostones of Unit B is sharp (Fig. 6B), suggesting that  
465 deposition of the mudstones postdated dolomitization of units A and B, 1 to 5 Ma. This contrasts  
466 with the surrounding bedrock of the Pedro Castle Formation where the boundary between the  
467 dolostone and limestone is gradational over a stratigraphic interval of ~ 2 m (MacNeil and Jones,  
468 2003, their Fig. 2).

469 *7.3. Package III (P-III)*

470 Package III is formed largely of red terra rossa that is locally intercalated with yellowish-  
471 brown flowstone (Fig. 6E). The terra rossa forms (1) laterally continuous laminae/beds (Fig. 4),  
472 (2) terra rossa breccia (Fig. 7A-D), and (3) ooids (Fig. 21). No marine biofragments are present  
473 in these deposits. This package, therefore, represents a significant change in character relative to  
474 the marine sediments found in the underlying sediment packages. The most obvious source for  
475 these sediments is the terra rossa soils found on the surface karst of Cayman Brac, as they are  
476 today. The distinctive flowstone, like that found in caves of all ages on the Cayman Islands,  
477 formed by precipitation from freshwater that flowed through the cave. U/Th dating of the  
478 flowstone indicates that it is more than 500,000 years old.

479 Although the poor soils of the Cayman Islands were mentioned in earlier studies (e.g.,  
480 Fawcett, 1888; Billymer, 1946), Ahmad and Jones (1969) were the first to provide some analyses

481 of the soils and Baker (1974) was the first to map the distribution of the different soils on the  
482 islands. Both Ahmad and Jones (1969) and Baker (1974) compared the red Cayman soils with  
483 the bauxites found on Jamaica. Ahmad and Jones (1969, their Table 1) showed that soils on  
484 Grand Cayman were formed largely of chlorite (36%), boehmite (27%), gibbsite (5%), kaolinite  
485 (1%), and amorphous  $\text{Al}_2\text{O}_3$  (5%),  $\text{SiO}_2$  (9%), and  $\text{Fe}_2\text{O}_3$  (15%). They argued that these soils  
486 differed from the Jamaican bauxites because they contained more silica but less alumina and  
487 iron. Later, Ahmad (1996, his Table 6) provided analyses of other soil samples, including two  
488 from Cayman Brac that were formed primarily of gibbsite, boehmite, kaolinite, and smectite  
489 along with lesser amounts of mica/illite, quartz, hematite, and goethite. Jones (1992a, his Table  
490 2), using data provided by the Water Authority, Cayman Islands showed that nine samples of  
491 terra rossa from the New Hut Farms area on the east end of Grand Cayman contained, on average  
492 (based on 9 samples), 20 ppm S, 22 ppm K, 1789 ppm Ca, 418 ppm Mg, 2 ppm Fe, 1 ppm Mn,  
493 <1 ppm Zn, < 1 ppm Cu, and 2 ppm Al. The Ca and Mg in these soils probably came from the  
494 dolostone bedrock.

495 The high calcite content in the terra rossa from the Upper Cave is due to the anatomizing  
496 arrays of calcite-filled microfractures (Fig. 14). The microfractures may have formed through  
497 desiccation of the terra rossa after it had been deposited in the caves. The calcite cements were  
498 precipitated from  $\text{CaCO}_3$  saturated waters that subsequently flowed through the cave.

499 The origin of the terra rossa breccia is difficult to explain because it is not a clast-supported  
500 breccia (Fig. 7A, B). The spaces between the terra rossa clasts are filled with a combination of  
501 flowstone and calcareous mudstone (Fig. 7A-D). The flowstone was precipitated from calcite-  
502 saturated meteoric waters. Although the source of the calcareous mudstone is unknown because  
503 no fossils were found in it, the O and C isotopes indicate that it was strongly influenced by

504 freshwater. The brecciated appearance of the terra rossa may be related to plant root activity.  
505 The best developed breccia is found close to the middle sinkhole, which may have once been the  
506 site where plants grew in terra rossa, as is commonly seen today on Cayman Brac and Grand  
507 Cayman. Evidence of root penetration is apparent at similar depths elsewhere in the cave (Fig.  
508 6A). The presence of roots would explain the separation of the clasts and would have also  
509 provided the channels for water flow that led to the precipitation of the flowstone.

510 The terra rossa ooids in the Upper Cave are similar to those found in the bauxite profiles in  
511 the Apennines of South Italy (Mondillo et al., 2011; Boni et al., 2013; Mongelli et al., 2014),  
512 Weipa in Australia (Tilley, 1998; Taylor and Eggleton, 2004; Eggleton et al., 2008; Taylor et al.,  
513 2008), the Darling Range of Australia (Anand and Paine, 2002; Anand and Verrall, 2011), and  
514 northern Saudi Arabia (Al-Mutairi et al., 2015). In many cases, the ooids are documented and  
515 illustrated without any comment on how they formed (e.g., Mondillo et al., 2011; Boni et al.,  
516 2013; Mongelli et al., 2014). Nadon (1991), however, argued that the Weipa pisoliths developed  
517 through glaebularisation or centripetal plasmic accumulation and alteration of preexisting  
518 kaolinite via Fe/Al oxihydration prior to formation of the cortices. Taylor and Eggleton (2004)  
519 suggested that that the cortices formed around fragments located in a porous weathered substrate  
520 and that many cracked as a result of mineral dehydration or desilicification. Although biological  
521 processes have generally been ignored, Anand and Verrall (2011) argued that pisoliths in the  
522 Darling Range (Australia) formed through the fungal-mediated precipitation of Al and Fe.  
523 Features documented for terra rossa ooids from other parts of the world and in the ooids found in  
524 the Upper Cave on Cayman Brac include nuclei of variable compositions that appear to be  
525 detrital in origin and cortical laminae that vary in appearance from ooid to ooid (Fig. 21). Some  
526 of the Cayman ooids are characterized by cortical laminae that are formed of calcite, and/or

527 mixtures of clays and calcite (Fig. 21). The Cayman ooids are held in calcite cement that was  
528 probably precipitated after the influx of clays and other terra rossa components had ceased (Fig.  
529 21).

#### 530 *7.4. Package IV (P-IV)*

531 P-IV includes the gastropod coquina (Fig. 7F) and the overlying bed of oncoid limestone  
532 (Fig. 23). Although found only in a small, high area of the cave that is located just north of the  
533 Middle Sinkhole (Fig. 4B), these facies are significantly different from the underlying facies.  
534 The scattered terra rossa lithoclasts (Fig. 7F) and terra rossa ooids (Fig. 23B, D) found in these  
535 facies were probably derived from the P-III sediments.

536 Given the uniformity of shell size and morphology, the gastropods found in the coquina  
537 appear to be a single species. Although probably of terrestrial origin, the specific species is  
538 unknown. The few shells that remain in the rock are well preserved and show little evidence of  
539 transportation. Gastropods and gastropod debris have been recorded from various caves  
540 throughout the world including Coco Ree, Jamaica (Goodfriend and Mitterer, 1993), Mé Auré,  
541 New Calidonia (Cowie and Grant-Mackie, 2004), and Mugnano Cave, Italy (Iacoviello and  
542 Martini, 2012, 2013). In an unnamed cave at Coco Ree, Jamaica, 40 species of land snails (600  
543 to 45,000 yrs B.P.), all endemic to Jamaica, were collected from unconsolidated or poorly  
544 cemented bauxitic sediments that overlie similar deposits that have been well-cemented by  
545 calcite (Goodfriend and Mitterer, 1993). All of the species found in the cave were also present  
546 on the exposed land surfaces around the cave. The setting is similar to that found in the Upper  
547 Cave in the West End Quarry on Cayman Brac. Similarly, Cowie and Grant-Makie (2004),  
548 documented 20 terrestrial species of gastropods from a cave at Mé Auré in New Caledonia that

549 had accumulated over a time span of ~ 3000 years. In each example, the gastropods appear to  
550 have been transported into the cave.

551 Many oncoids in the bed above the gastropod coquina, have small gastropod shells or  
552 fragments of larger shells as their nucleus (Fig. 23). Although the exact conditions under which  
553 they formed is difficult to interpret from the limited evidence available, it was probably a high  
554 energy setting with water more or less constantly flowing through the area.

#### 555 *7.5. Package V (P-V)*

556 P-V includes the flowstone in the upper part of the cave that lies on top of the terra rossa  
557 breccia (P-III), the stalactites and draperies that adorn the cave ceiling close to the Middle  
558 Sinkhole (Fig. 7D, E), and rare small stalagmites. These deposits formed from waters that  
559 flowed over the cave floor (flowstone) and dripped from the ceiling (stalactites). The calcite  
560 cements that filled the microfractures in the underlying terra rossa (Fig. 14), the flowstone that  
561 formed around the terra rossa lithoclasts (Fig. 7A-C), and locally on top of the red terra rossa  
562 (Fig. 7D) were probably formed from the same waters.

563 U/Th dating of a flowstone/stalagmite sample from this package yielded ages of >500,000  
564 years (lower flowstone), 64,355 years (lower stalagmite), and 22,500 and 26,353 years old. Such  
565 dates attest to the antiquity of the cave and the shows that such precipitation was episodic.

### 566 **8. Geological evolution of late-stage cavities**

567 Irregular shaped cavities that seem to extend from the present day surface, locally crosscut  
568 the bedrock and the deposits in both the Lower Cave and Upper Cave. These cavities are filled  
569 with red terra rossa or olive terra rossa. As such, their formation must post-date development of  
570 those caves and most of the deposits that fill them. The cavities filled with the olive terra rossa

571 are younger than those filled with the red terra rossa because the former commonly crosscut the  
572 latter.

## 573 **9. Discussion**

574 Well-developed karst terrains with spectacular arrays of caves are common in the carbonate  
575 bedrock of many isolated oceanic islands, including those found in the Pacific Ocean (e.g.,  
576 Rodrigues Island – Burney et al., 2015), the Mediterranean Sea (e.g., Mallorca – Fornós et al.,  
577 2009; Fornós et al., 2014), the North Atlantic Ocean (e.g., Bermuda – Palmer et al., 1977;  
578 Mylroie, 1984; van Hengstum and Scott, 2012), and the Caribbean Sea (e.g., the Bahamas –  
579 Mylroie, 1984; Mylroie et al., 1991). If the caves are partly or completely submerged by  
580 seawater they become receptacles for marine sediments (e.g., Fornós et al., 2009; Farrant and  
581 Smart, 2011; van Hengstum and Scott, 2012), whereas those in the vadose zone may be  
582 characterized by marine sediments, terrestrial sediments, and/or speleothems (e.g., Auler et al.,  
583 2009; Farrant and Smart, 2011; van Hengstum et al., 2011; van Hengstum and Scott, 2012). In  
584 these dynamic settings, variations in sea level, ~~be they due to eustasy or tectonism~~, will cause a  
585 reset of the entire system. Thus, a drop in sea level may place a previously submerged cave in  
586 the vadose zone whereas a rise in sea level may lead to submergence of a cave that had been in  
587 the vadose zone. Potentially, the deposits found in the caves will provide a record of such  
588 changes, which may not be evident from the host bedrock succession. Such is the case for caves  
589 developed in the Miocene and Pliocene carbonates on Cayman Brac.

590 **During the late Pliocene and Pleistocene**, Cayman Brac was subject to rapid eustatic  
591 oscillations in sea level (Fig. 25). Establishing the timing and scale of the eustatic sea level  
592 changes is difficult given the contrasting opinions that exist. For example, interpretations of  
593 highstand levels during the Mid-Pliocene Warm Period (~3.3 to 2.9 Ma), based on physical

594 features, have yielded estimates of + 35 m from North and South Carolina (Dowsett and Cronin,  
595 1990), +15-20 m from Virginia (Krantz, 1991), +20-25 m from Enewetak Atoll (Wardlaw and  
596 Quinn, 1991), +60 m from Alaska (Kaufman and Brigham-Grette, 1993), and +30 m from Roe  
597 Plain, Australia (James et al., 2006). This gives an overall range of +15 to 60 m with an average  
598 of ~ 34 m. Raymo et al. (2011), however, argued that these estimates were too high because they  
599 had not considered all of the factors that have led to the present-day altitudes of the physical  
600 features on which these sea levels were based. Their modeling for these areas, which included  
601 corrections for (1) local tectonic movements, (2) local sediment loading, (3) changes due to  
602 mantle convection flow, and (4) glacial isostatic adjustments reduced the estimated sea-level  
603 highstands to +7.9 to 26.8 m (Raymo et al., 2011). Comparison of sea-level curves like those  
604 shown in Figure 2A (Miller et al., 2005) and Figure 25B (Hansen et al., 2013), also highlight the  
605 contrasts between different models. Thus, caution must be used in assessing the succession on  
606 Cayman Brac relative to eustatic sea level variations.

607 For Cayman Brac, the situation is further complicated because the exact timing of the  
608 tectonic tilting to the west is unknown (Zhao and Jones, 2012, 2013; Liang and Jones, 2015a).  
609 Two models (I and II) have been invoked to explain the evolution of the bedrock succession,  
610 with the key difference being the timing of the tectonic tilting (Zhao and Jones, 2012).

- 611 • Model I involves (1) deposition of the Cayman Formation followed by development of the  
612 Cayman Unconformity, and later dolomitization, (2) tectonic tilting of the island to the west,  
613 and (3) deposition of the Pedro Castle Formation only on the western part of the island.
- 614 • Model II involves (1) deposition of the Cayman Formation, (2) development of the Cayman  
615 Unconformity, (3) deposition of the Pedro Castle Formation over the entire island, (4)

616 tectonic tilting of the island, (5) removal, by erosion, of all of the Pedro Castle Formation  
617 from the eastern and central parts of the island.

618 Based on available evidence, Zhao and Jones (2012) adopted Model II and thereby argued  
619 that tilting started after the sediments of the Pedro Castle Formation had been deposited. Tilting  
620 must have ended prior to deposition of the sediments of the Ironshore Formation, 125,000 years  
621 ago, because the associated wave-cut notch (6 m asl) evident in the cliff faces around the island,  
622 is horizontal and crosscuts the westward dipping strata of the Cayman Formation and Pedro  
623 Castle Formation.

624 Caves are common on Cayman Brac (Gilleland, 1998), including many with their entrances  
625 clearly evident in the cliff faces (Tarhule-Lips and Ford, 1998, 2004). Divided into the “notch  
626 caves” (entrances < 2 m above notch) and the “upper caves” (entrances > 2 m above notch), they  
627 are characterized by speleothems up to 408,000 years old but little clastic sediment (Tarhule-Lips  
628 and Ford, 2004). The notch caves, with some containing speleothems > 200 ka, must have  
629 formed before the +6 m notch that developed ~125 ka (Tarhule-Lips and Ford, 2004). Caves  
630 located on top of the bluff, inland from the cliff faces, are typically small and decorated with a  
631 variety of speleothems (Jones, 2010). Flowstone in the lower part of the Upper Cave in West  
632 End Quarry is > 500,000 years old, indicating that some of these caves predated those evident in  
633 the cliff faces.

634 The Lower Cave, housed in the Cayman Formation on Cayman Brac is similar to a filled  
635 cave that once existed (now destroyed by blasting) in the Cayman Formation in Pedro Castle  
636 Quarry on Grand Cayman that had its floor ~ 9 m below the Cayman Unconformity (Jones and  
637 Smith, 1988, their Fig. 6A, B). A tunnel from its west end connected the cave to a large sinkhole  
638 that had its opening at the Cayman Unconformity (Jones, 1992b, his Fig. 2). The cave was filled

639 with caymanite, dolomitized wackestone (with marine fossils), red terra rossa, and flowstone  
640 (Jones, 1992b, his Fig. 2). Emplacement of the red terra rossa and flowstone postdated the Pedro  
641 Castle Formation and dolomitization. The caymanite and marine sediments found in the caves  
642 on Cayman Brac and Grand Cayman indicate that both cave entrances must have been open to  
643 the influx of the marine sediments. Those sediments must have emplaced prior to deposition of  
644 the sediments that now formed the Pedro Castle Formation. As such, both caves had similar  
645 developmental histories.

646 The diverse array of cave-filling sediments and precipitates in the Upper Cave in the West  
647 End Quarry on Cayman Brac reflect the complex evolutionary history of that cave. Evident from  
648 these deposits (P-I to P-V) are three key evolutionary stages.

- 649 • Restriction of dolomitization to P-I. Zhao and Jones (2012) argued that Phase II  
650 dolomitization took place while the sediments that now form the Pedro Castle Formation  
651 were being deposited. Given that the dolostones of P-I occur in a cave located in the  
652 lithified strata of the Pedro Castle Formation, it follows that their dolomitization must have  
653 postdated the Phase II dolomitization of Zhao and Jones (2012). The exact age of this late  
654 phase dolomitization is, however, impossible to determine because the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of the  
655 P-I dolostones fall within the range of ratios obtained from the dolostones of the Cayman  
656 Formation and Pedro Castle Formation (Fig. 24).
- 657 • Transitional conditions of P-II. The lack of dolomite in these mudstones suggests that  
658 deposition postdated dolomitization and the presence of foraminifera in the mudstones  
659 indicates that the cave was open to marine incursions with its entrance close to sea level.  
660 The negative  $^{18}\text{O}$  and  $^{13}\text{C}$  values of the mudstone, which are lower than those of the

661 underlying dolostones of P-I, indicate that meteoric waters were involved in their  
662 formation.

- 663 • With the onset of P-III all connection to the marine environment was lost as shown by the  
664 lack of marine sediments in P-III, P-IV, and P-V and the negative  $^{18}\text{O}$  and  $^{13}\text{C}$  signatures of  
665 those deposits (Fig. 24). Package III involve the first precipitation of flowstone and the  
666 first influx of terrestrial sediments (Figs. 24, 25) as surface soils (terra rossa) were washed  
667 into the cave. U/Th dating indicates that the flowstone in the base of this package is >  
668 500,000 years old, whereas the uppermost speleothems in P-IV are only  $\sim 21,000$  years old.

669 Development of the notch, upper, and top caves suggests that the core of the island has  
670 probably remained above sea level for at least the last 500,000 years and certainly ever since  
671 deposition of the sediments that now form the Pedro Castle Formation ended. Since then the  
672 uplifted core of the island has been actively eroded. Evidence presented by Liang and Jones  
673 (2015a), for example, suggests that as much as 145 m of the Cayman Formation and 45 m of the  
674 Pedro Castle Formation have been lost to erosion at East Point, which is  $\sim 16.3$  km to the ENE of  
675 the West End Quarry. In the quarry, only the basal 2-3 m of the Pedro Castle Formation  
676 remains, meaning that 42-43 m of the formation has been lost to erosion and only the oldest part  
677 of the formation remains. Today, the surface of the uplifted core, formed of the Cayman  
678 Formation and the Pedro Castle Formation (westernmost part of island only), is characterized by  
679 a rugged karst terrain (e.g., Liang and Jones, 2015a, their Fig. 11) with terra rossa accumulations  
680 in many of the depressions and sinkholes (e.g., Ahmad, 1996).

681 The red terra rossa, the most obvious deposit in the Upper Cave (Fig. 4), is significant  
682 because it is the first record of terrestrial sediments in the cave. Many of the younger caves on  
683 Cayman Brac have “red soils” on their floors that came from the terra rossa soils found on the

684 surface (Tarhule-Lips and Ford, 2004). Although the subject of much debate, Muhs et al. (2007)  
685 argued that soils found on limestone islands in the western Atlantic Ocean and Caribbean Sea are  
686 formed of (1) insoluble residues derived from the carbonate bedrock, (2) fluvial transport of  
687 clays from nearby siliciclastic outcrops, (3) wind-blown dust, and/or (4) volcanic ash. For  
688 Cayman Brac, option 2 can be excluded because there are no siliciclastic rocks on the island.  
689 Although option 4 is viable, available evidence indicates that volcanic input on Cayman Brac  
690 was minimal. The possibility that the soils are formed of insoluble residues is viable given that  
691 erosion has removed as much 190 m of limestones/dolostones from parts of Cayman Brac. The  
692 limestone and dolostone bedrocks of Cayman Brac, however, are pure and it is doubtful that the  
693 loss of 190 m of limestone/dolostone could have produced the volume of soil now found on the  
694 island. This notion parallels the conclusions offered by Tracey et al. (1964), Birkeland (1999),  
695 and Muhs et al. (1987) for soils found on top of carbonate strata on other islands. It appears,  
696 therefore, that most of the soils found on the Cayman Islands were derived largely from wind-  
697 borne dust from the Sahara Desert and/or from North America during the last glacial period  
698 when winds from the west and northwest transported loess over the Caribbean Sea (Muhs et al.,  
699 2007).

700 Today, the floor of the Upper Cave on Cayman Brac is ~ 5 m asl. Stratigraphic evidence  
701 suggests that this cave probably formed over the last 2.0 to 4.5 million years and it must have  
702 postdated lithification of the Pedro Castle Formation (Fig. 25). Nevertheless, the precise age for  
703 the onset of cave formation is impossible to determine given that the exact age of the uppermost  
704 part of the Pedro Castle Formation on Cayman Brac is unknown because erosion has removed  
705 everything but the basal part of the formation. Thus, it is impossible to determine if

706 sedimentation was restricted to the lower part of the Pliocene or if it continued throughout the  
707 Pliocene.

708         In the Upper Cave, the base of P-III (~ 5.3 m asl) is significant because it records the time  
709 when the cave became isolated from marine influences and was first emplaced in the vadose  
710 zone (Fig. 25). That change, however, might have been due to eustatic changes in sea level  
711 and/or tectonic uplift of the island. Hansen et al. (2013) suggested that sea levels were +10-20 m  
712 throughout most of the Pliocene (Fig. 25B), which would have allowed sediment deposition of  
713 the sediments that now form the Pedro Castle Formation but precluded formation of the Upper  
714 Cave. This sea level curve also suggests that it was not until the latest Pliocene that sea levels  
715 dropped below present day sea level (Fig. 25B). This would have led to exposure of the Pedro  
716 Castle Formation and the formation of karst, which would have continued throughout the  
717 Pleistocene apart from short-lived highstands (Fig. 25B). If the succession in the Upper Cave is  
718 considered relative to this sea level curve, then it could be speculated that the base of P-III is  
719 roughly coincident with the Pliocene-Pleistocene boundary. This attractive proposition,  
720 however, implicitly assumes that the sea level curve proposed by Hansen et al. (2013) is correct,  
721 and ignores the fact that tectonic uplift and tilting was also taking place during this time period.

722         Tectonic elevation and tilting of the core of Cayman Brac could also have been responsible  
723 for emplacement of the Upper Cave into the vadose zone with its entrance beyond the influence  
724 of marine waters. Uplift would probably may also have caused fracturing and faulting in the  
725 bedrock (cf., Purdy and Waltham, 1999) that, in turn, promoted water circulation and breakdown  
726 of the exposed Pedro Castle Formation and Cayman Formation. As uplift proceeded, erosion  
727 progressively removed the Pedro Castle Formation and much of the Cayman Formation from the  
728 central and eastern parts of the island.

729           With the information presently available it is impossible to determine if development of the  
730 bedrock succession, the caves, and the cave fills was controlled by eustatic sea level changes,  
731 tectonic tilting, or some combination of the two. Nevertheless, it is readily apparent that  
732 integration of the information derived from the bedrock succession and the cave-filling  
733 successions has provided insights into the geological evolution of Cayman Brac that cannot be  
734 determined from the bedrock alone.

## 735 **10. Conclusions**

736           The development of limestone successions on isolated oceanic islands is fundamentally  
737 controlled by the interplay between eustatic sea level changes and local tectonic activity.  
738 Deposition during the highstands leaves a tangible record of the operative processes, whereas  
739 lowstands typically leads to karst formation as dissolution modifies the bedrock. Information on  
740 other associated processes, including sediment formation or precipitation, can be difficult to  
741 ascertain. This shortcoming, however, can be partly overcome by interpretation of deposits  
742 found in caves. This is well illustrated in this study where detailed analysis of cave-filling  
743 deposits in the Miocene-Pliocene strata of Cayman Brac has led to the following major  
744 conclusions.

- 745       • The Lower Cave and the caymanite that fills it developed during the Messinian lowstand.  
746       Marine fossils in the caymanite indicate that the cave entrance was open to marine  
747       incursions, possibly during storms.
- 748       • The Upper Cave is filled with various lithotypes that developed in response to ever  
749       changing conditions. The basal sediment packages in this cave formed under marine  
750       influences whereas the upper sediment packages were isolated from marine incursions and  
751       developed in a meteoric regime.

- 752 • U/Th dating indicates that some of the flowstones are > 500,000 years old whereas other  
753 flowstone in the uppermost part of the Upper Cave formed ~ 62,000 and 21,000 years ago.
- 754 • Late stage cavities that cut across both the Lower Cave and the Upper Cave are filled with  
755 red and olive terra rossa with the latter being younger than the former.

756 Integration of these conclusions with interpretations of the bedrock succession provides  
757 valuable information about the subaerial landscape as it evolved while the core of the island  
758 remained above sea level.

759

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765 End Quarry on Cayman Brac; Diane Caird who did the XRD analyses; Noga Vaisblat who  
766 processed the ion-milling of the terra rossa sample; and Nathan Gerein who operated the SEM  
767 used in this study.

768

768

**FIGURE CAPTIONS**

769 **Fig. 1.** Location of West End Quarry on Cayman Brac. (A) Location of Cayman Brac in  
770 Caribbean Sea. (B) Geological map of Cayman Brac showing location of West End Quarry.  
771 Map adapted from Jones (1994). (C) Geological cross-section through Cayman Brac (from X  
772 to Y on panel B). Adapted from Jones (1994).

773 **Fig. 2.** (A) Stratigraphic succession on west end of Cayman Brac. (B) Sea-level curve for last 10  
774 Ma based on data in Miller et al. (2005, Supplemental Table S1 with points plotted every  
775 0.05 Ma interval). Dates for Messinian Salinity Crisis (MSC) onset and termination taken  
776 from Jiménez-Moreno et al. (2013). Dates for the periods based on the International  
777 Chronostratigraphic Chart  
778 (<http://www.stratigraphy.org/ICSchart/ChronostratChart2014-02.jpg>). Modified from  
779 Liang and Jones (2015, their Fig. 2).

780 **Fig. 3.** View of northwest corner West End Quarry showing the Cayman Formation (CF),  
781 Cayman Unconformity (CUC), Pedro Castle Formation (PCF), and modern karst surface.  
782 Note open caves and cavities (C) filled with lithified terra rossa of various colours.  
783 Photograph taken in 2006 when quarry floor and top of quarry face were ~ 1.5 m and ~ 8 m  
784 above sea level, respectively.

785 **Fig. 4.** East wall of West End Quarry showing cross-sections through filled caves. (A) North  
786 end of Lower Cave (outlined by black line) in the Cayman Formation (CF), the Cayman  
787 Unconformity, the south end of the Upper Cave in the Pedro Castle Formation (PCF), and the  
788 “South Sinkhole”. (B) Central and north parts of Upper Cave (outlined by black line) and  
789 location of “Middle Sinkhole”. North end of the cave is not shown because poor outcrop  
790 conditions prevents detailed mapping. Red arrow is at same point as red arrow on panel A.

791 **Fig. 5.** Lower Cave in the Cayman Formation (Fig. 4A). (A, B) Caymanite with red and off-  
 792 white laminae defining variable attitudes of the laminae. CF= Cayman Formation. (B, C)  
 793 Cavities cross-cutting the caymanite and filled with olive terra rossa (OTR) and red terra  
 794 rossa (RTR).

795 **Fig. 6.** Outcrop photographs of deposits in basal part of the Upper Cave housed in the Pedro  
 796 Castle Formation (Fig. 4A). Panels A-D from south end of cave; panel E from middle part of  
 797 the cave. CF = Cayman Formation, PCF = Pedro Castle Formation, RTR = red terra rossa,  
 798 OTR = olive terra rossa, CM = calcareous mudstone; F = flowstone, SC = spar calcite. (A)  
 799 General view showing base of Upper Cave at the Cayman Unconformity (arrow). Cave fill is  
 800 divided into units A and B (dolostones), and units C and D (limestone and terra rossa). PD =  
 801 location of image shown in panel D. (B) Basal parts of Upper Cave showing sharp boundary  
 802 between the dolostone of Unit B and calcareous mudstones of Unit C. (C, D) Unit C formed  
 803 of white to red calcareous mudstones and Unit D formed of irregular masses of red terra  
 804 rossa in a limestone groundmass. Note isolated cavities filled with red and olive terra rossa.  
 805 (E) Basal part of Upper Cave with flowstone intercalated with red calcareous mudstone and  
 806 red terra rossa.

807 **Fig. 7.** Outcrop photographs of deposits in middle and upper parts of the Upper Cave near the  
 808 Middle Sinkhole (Fig. 4B). RTR = red terra rossa, S = speleothem, LST = limestone. (A)  
 809 Terra rossa breccia formed of irregular-shaped masses of red terra rossa held in groundmass  
 810 formed of speleothemic calcite and limestone. Note open cavities. (B) Enlarged view from  
 811 lower right part of panel A showing showing spaces between the masses of red terra rossa  
 812 filled with speleothemic calcite and limestone. (C) Enlarged view showing red terra rossa  
 813 coated with thinly laminated speleothemic calcite and central cavity filled with limestone.

814 (D) Upper part of cave filled with terra rossa breccia overlain by speleothemic calcite and  
 815 stalactites and draperies hanging from cave roof. PE indicates drapery shown in panel E. (E)  
 816 Ornate drapery hanging from cave roof. (F) Uppermost part of cave fill formed of gastropod  
 817 shells embedded in matrix formed of coated grains held in spar calcite cement.

818 **Fig. 8.** Thin section photomicrographs of caymanite (all dolomite) from Lower Cave. (A)

819 Laminae defined by subtle variations in colour and grain sizes. (B) Grainstone laminae  
 820 overlying mudstone laminae. (C) Small red algae biofragments in grainstone laminae. (D)  
 821 Planktonic foraminifera in mudstone laminae.

822 **Fig. 9.** SEM photomicrographs of caymanite from Lower Cave. (A) Finely crystalline dolomite

823 groundmass with small pores lined with limp dolomite (LD). (B) Caymanite from upper  
 824 part of cave, formed of very finely crystalline dolomite (left side) with pore lined by limp  
 825 dolomite (LD) and filled with calcite (C).

826 **Fig. 10.** SEM photomicrographs of finely crystalline dolostones from units A (A-C) and B (D-F)

827 in Upper Cave (Fig. 6A). (A) Finely crystalline dolostone with pore filled with calcite (C).  
 828 (B) Enlarged view of dolostone showing interlocking dolomite crystals. (C) Dolomitized red  
 829 algae fragment held in dolostone matrix. (D) Finely crystalline dolostone from Unit B. (E)  
 830 Enlarged view of dolostone showing interlocking euhedral dolomite crystals. (F) Hollow  
 831 dolomite crystals, Unit B.

832 **Fig. 11.** Thin section photomicrographs of calcareous mudstones from Unit C in Upper Cave

833 (Fig. 6A). Images A-D, unstained thin section; E-G stained with Alizarin Red S Solution. (A)  
 834 White mudstone with microfractures and cavities lined with calcite cement. (B) Graded  
 835 bedding in dipping pink mudstone laminae. (C, D) Examples of foraminifera in calcareous  
 836 mudstones. (E) Microbrecciated calcareous mudstone with calcite cement. (F) Small cavity

837 with small clasts at base, each surrounded by calcite cement, overlain by mudstone that is  
 838 coated with calcite cement. (G) Fracture through mudstone lined with calcite cement and  
 839 filled with olive terra rossa.

840 **Fig. 12.** SEM photomicrographs of calcareous mudstones from Unit C. (A, B) General and  
 841 enlarged views of white mudstone. (C) Red mudstone with small cavity filled with spar  
 842 calcite cement. (D) Red mudstone with disseminated patches of Fe-rich precipitates (arrows).  
 843 (E) Enlarged view of Fe-rich mass covered with interlocking plate-shaped crystals. Point A –  
 844 EDS analysis (see Fig. 13 A). (F) Enlarged view of plate-shaped crystals from panel E. Area  
 845 B – EDS analysis (see Fig. 13B). (G) Rounded mass of Fe-rich precipitate, from off-white  
 846 mudstone, formed of trigonal-shaped subcrystals. Point C – EDS analysis (see Fig. 13C). (H)  
 847 Series of linked Fe-rich precipitates in off-white calcareous mudstone. I indicates position of  
 848 image shown in panel I. (I) Enlarged view of Fe-precipitate. Point D – EDS analysis (see Fig.  
 849 13D). (J) Two ovate masses of Fe-precipitates, each formed of rounded subcrystals. Point E –  
 850 EDS analysis (see Fig. 13E). (K) Agglomeration of small Fe-rich precipitates in white  
 851 mudstone. L indicates position of image shown in panel L. (L) Fe-rich precipitate formed of  
 852 rounded subcrystals. Point F – EDS analysis (see Fig. 13F).

853 **Fig. 13.** EDS analyses for Fe-rich precipitates found in calcareous mudstones shown in Figure  
 854 12. Weight percentages of elements determined from EDS analyses.

855 **Fig. 14.** Thin section and BSE images of red terra rossa. (A-C) Thin section photomicrographs  
 856 of terra rossa (yellow to orange) dissected by anatomizing arrays of calcite-filled  
 857 microfractures. (D, E) BSE images of polished surface showing contrast between red terra  
 858 rossa (dark grey) and calcite-filled fractures (light grey). (F) Elemental map of surface shown  
 859 in panel E (Ca and Al are in same positions on both panels) showing distribution of Al

860 (yellow) relative to calcite (red). Al-rich areas also contain Si and Fe. (G) Thin section  
 861 photomicrograph showing bands of yellowish calcite between bands of red terra rossa. (H)  
 862 BSE image of same area as in panel G, showing contrast between the calcite (light grey) and  
 863 red terra rossa (dark grey). Line X-Y indicates position of line transect shown in Figure 15.  
 864 (I) Enlarged view from lower part of panel G showing alternating bands of fibrous calcite  
 865 and red terra rossa.

866 **Fig. 15.** Compositional transect along line X-Y (Fig. 14H), showing contrast between the calcite  
 867 and red terra rossa. Minor elements, not shown in this diagram are Ti, Mg, and K (~ 4 wt%  
 868 in total), are found largely in the red terra rossa. Based on 100 equally-spaced points of  
 869 analysis.

870 **Fig. 16.** SEM photomicrographs of micro-milled surface of red terra rossa. Elemental content  
 871 based on EDS analyses. (A-C) Groundmass of lath-shaped crystals (Al and Si) of two size-  
 872 classes with disseminated Fe-oxide grains (Fe). (D) Mass of Fe-oxide in terra rossa matrix.  
 873 (E) Disseminated Fe-oxide grains in terra rossa matrix. (F) Euhedral zircon crystal (Zr and  
 874 Si). (G) Ti-rich grain. (H) Grain formed of Cr and Fe. (I) Grain formed of P, Ce, and La.

875 **Fig. 17.** Thin section photomicrograph (A) and BSE images (B-E) of terra rossa in terra rossa  
 876 breccia. (A) Contrasting colours of terra rossa evident in thin section. (B) BSE image of  
 877 central part of panel A (white arrow indicates common point) showing calcite-filled (Ca)  
 878 fractures and lack of contrast between the different coloured terra rossa. (C) Enlarged view  
 879 from panel B (white arrow indicates common point) showing lack of contrast between  
 880 different coloured terra rossa and location of transect shown in Figure 18. D and E indicate  
 881 location of areas shown in panels D and E. (D, E) Comparison of disseminated Fe (white) in  
 882 different coloured terra rossa.

883 **Fig. 18.** EDS analyses along transect U-V (Fig. 17C). For ease of comparison, the weight % of  
884 Al, Si, Fe, and Ti have been added together (ASFT) given that they are always associated  
885 with each other in the terra rossa. Based on 100 equally spaced points of analysis.

886 **Fig. 19.** Thin section photomicrographs (A, B, D) and BSE images (C, E) of olive terra rossa.  
887 (A) Variations in colour of olive terra rossa as seen in plane polarized light. Letters B and D  
888 indicate locations of panels B and D, respectively. (B) Enlarged view of bright red area of  
889 terra rossa (panel A) showing subtle colour variations. (C) BSE image of same area as panel  
890 B and location of transect W-X (Fig. 20A). (D) Enlarged view of terra rossa (panel A) with  
891 array of calcite-filled microfractures. (E) BSE image of same area shown in panel D and  
892 location of transect Y-Z (Fig. 20B). Calcite-filled microfractures highlighted by light grey  
893 colour.

894 **Fig. 20.** EDS line transects along lines W-X (Fig. 19C) and Y-Z (Fig. 19E). Each transect is  
895 based on 100 equally spaced points of analysis. Note difference in Ca levels between the two  
896 transects.

897 **Fig. 21.** Paired thin section photomicrographs (top) and BSE images (bottom) showing variations  
898 in structures and composition of terra rossa ooids from Unit D (Fig. 6C). Black-white-yellow  
899 lines on BSE images show EDS transects for compositional analyses shown in Figure 22.  
900 Black = line of transect; white = nucleus of ooid; yellow = cortical laminae and correspond to  
901 zones indicated on transects in Figure 22. Ooids held in calcite groundmass.

902 **Fig. 22.** EDS analyses showing contrast in distribution of calcium (C) and Al+Si+Fe+Ti (ASFT)  
903 of terra rossa ooids shown in Figure 21. Dashed lines indicate boundaries between the nuclei  
904 (N) and cortical laminae (C) of ooids as indicate in Figure 22. Each transect is based on 100  
905 equally spaced points of analysis.

906 **Fig. 23.** Thin section photomicrographs of coated grains from uppermost part of Upper Cave  
907 near the Middle Sinkhole. All images with plane polarized light. Blue = porosity. (A, B)  
908 Gastropod shells coated with thin micrite laminae and isopachous calcite rim. Interiors partly  
909 filled with terra rossa ooids and calcite cement. (C) Group of small coated grains, each with  
910 shell fragment as its nucleus. (D) Enlarged view of uppermost coated grain from panel C  
911 showing nucleus formed of shell fragment and successive layers of micrite highlighted by  
912 subtle differences in colour. (E) Group of coated grains with nuclei formed of shell fragments  
913 and encased by isopachous calcite cement. Note small terra rossa ooids between the coated  
914 grains. (F) Coated grain formed large micrite nucleus encased by micrite layers.

915 **Fig. 24.** Schematic stratigraphic column (not to scale) showing (A) bedrock succession, the  
916 Lower Cave, and cave deposits in the Upper Cave that are divided into genetically related  
917 packages P-I to P-V. (B)  $^{18}\text{O}$  and  $^{13}\text{C}$  isotopes for different lithotypes found in the caves  
918 (circles) relative to the ranges of values (bars) for the Cayman Formation (Zhao and Jones,  
919 2012) and Pedro Castle Formation (from MacNeil and Jones, 2003). (C)  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios for  
920 the dolostones in the lower and Upper Caves (squares) relative to the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios for the  
921 dolostones (bars) in the Cayman Formation and Pedro Castle Formation (MacNeil and Jones,  
922 2003; Zhao and Jones, 2012).

923 **Fig. 25.** Integration of paragenetic stages in evolution of bedrock succession (A) and cave-fill  
924 succession (C) relative to sea-level changes over the last 5.33 myrs (from Hansen et al.,  
925 2013, their Fig. 2b). Pliocene-Pleistocene boundary is placed at 2.58 myrs rather than at 1.8  
926 myrs as shown by Hansen et al. (2013).

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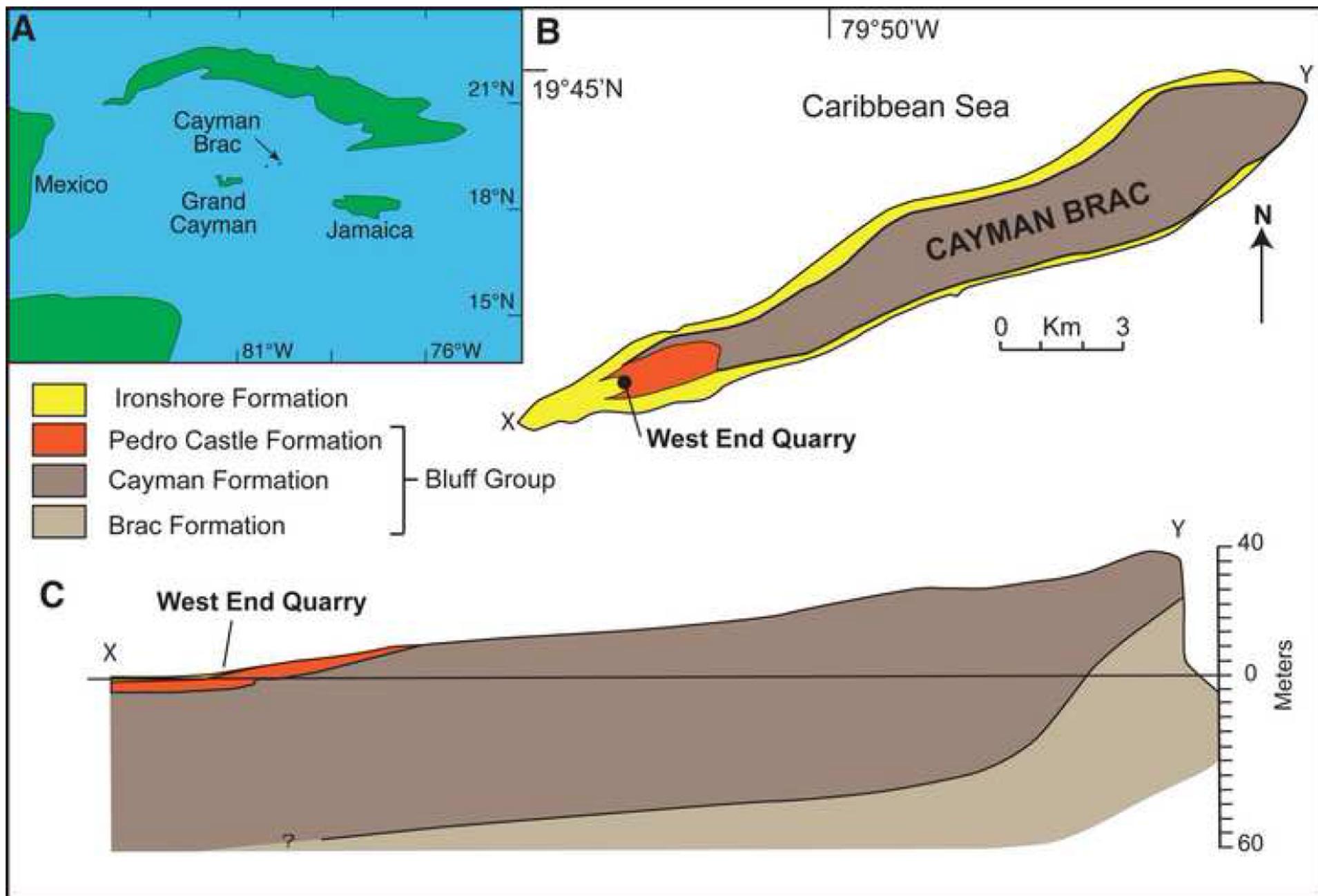


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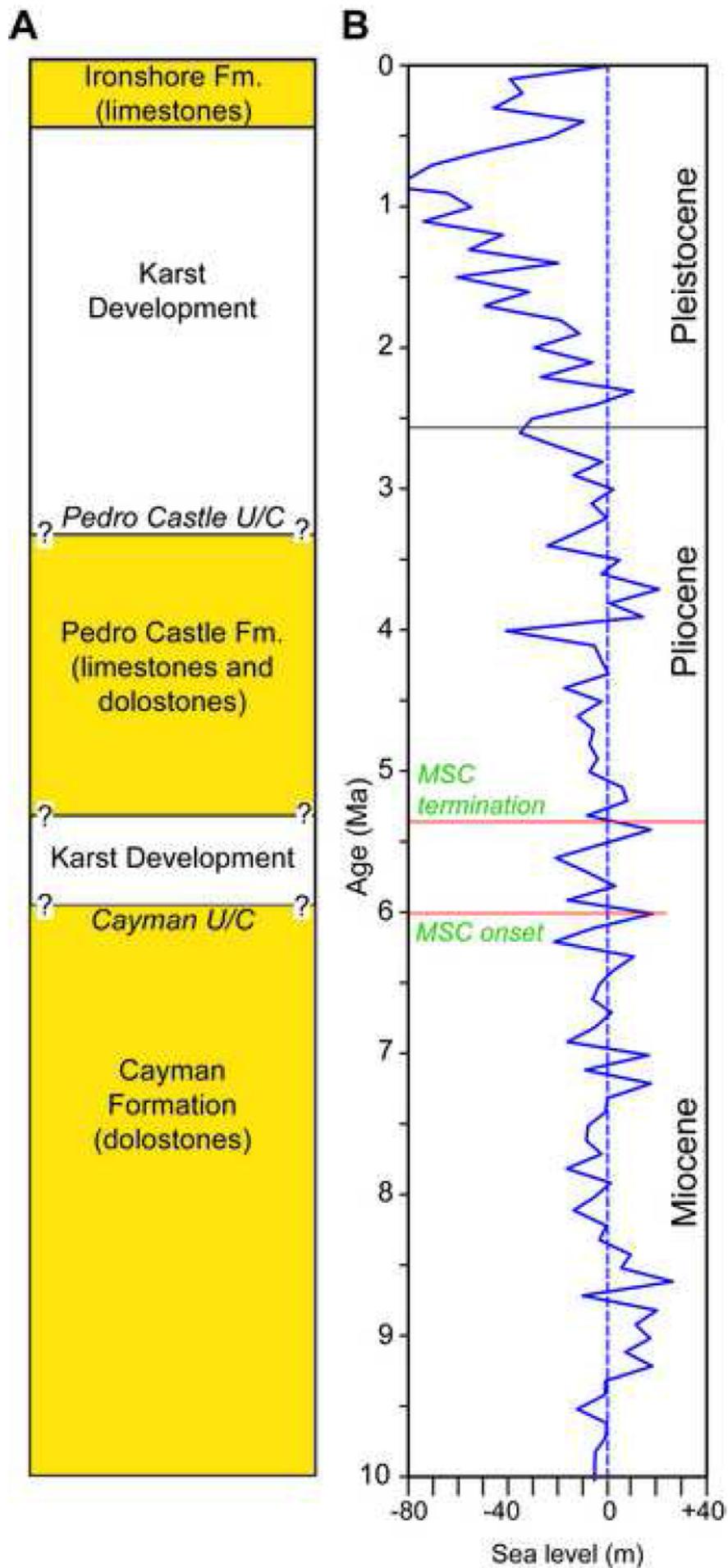


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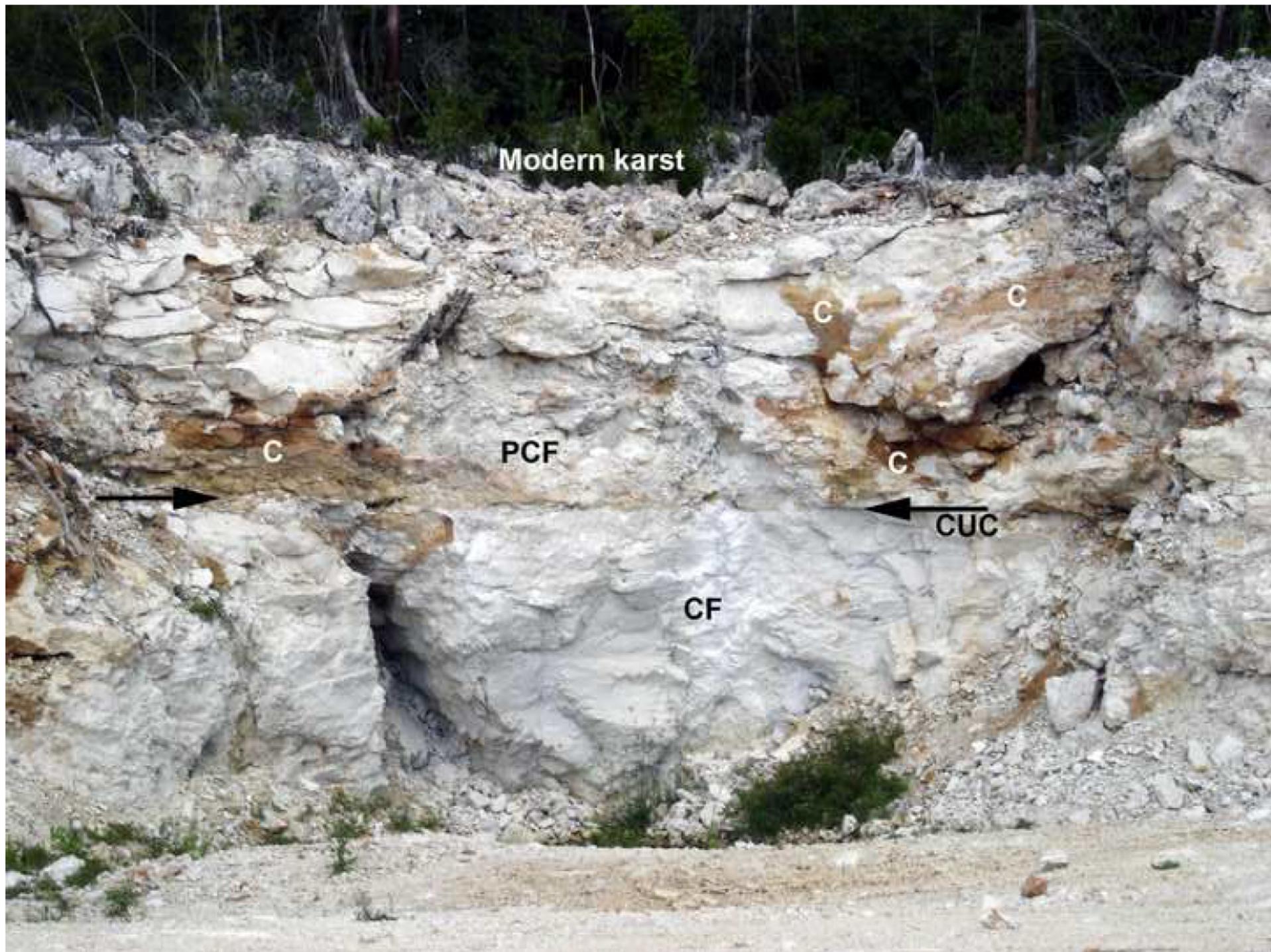


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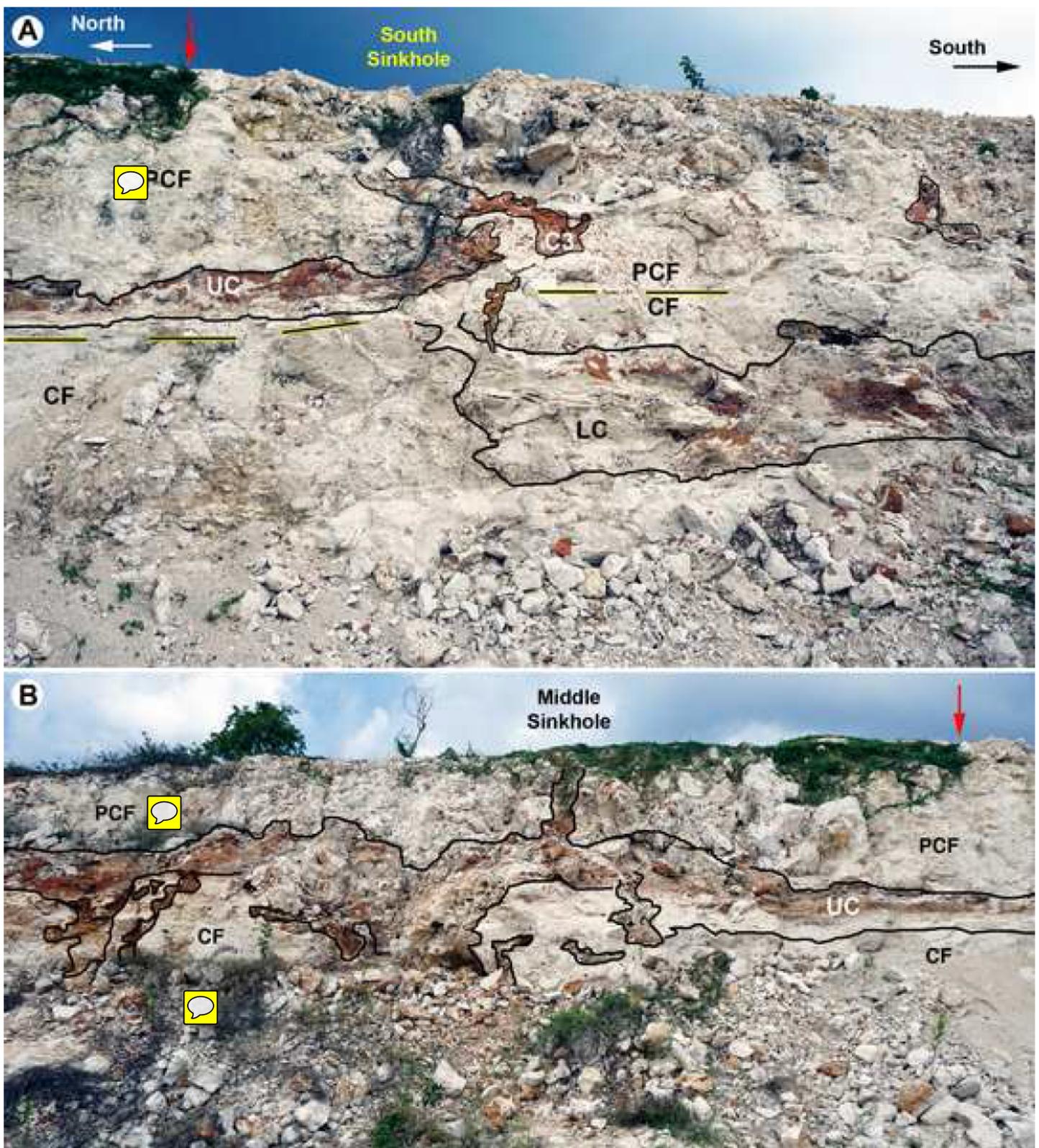


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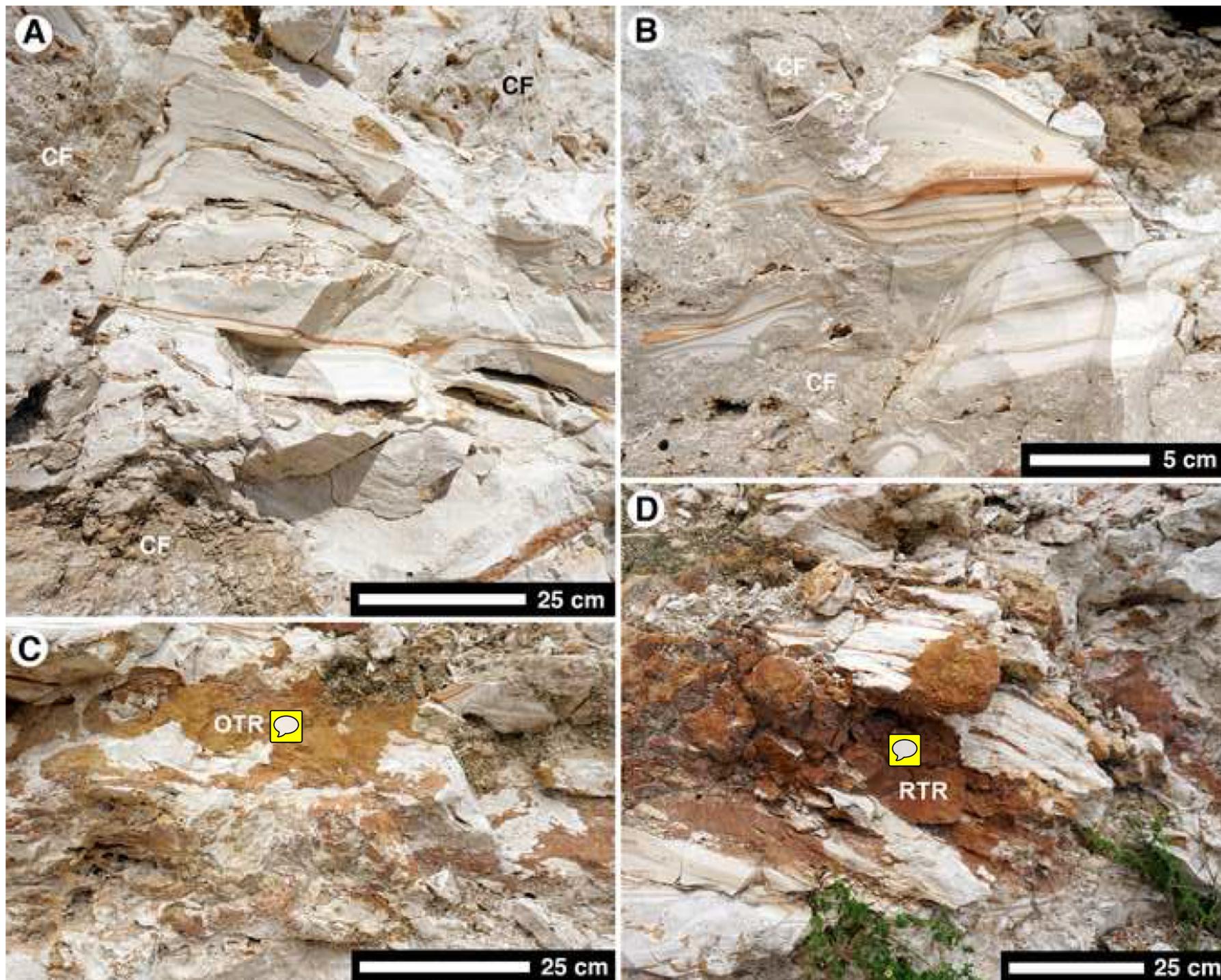


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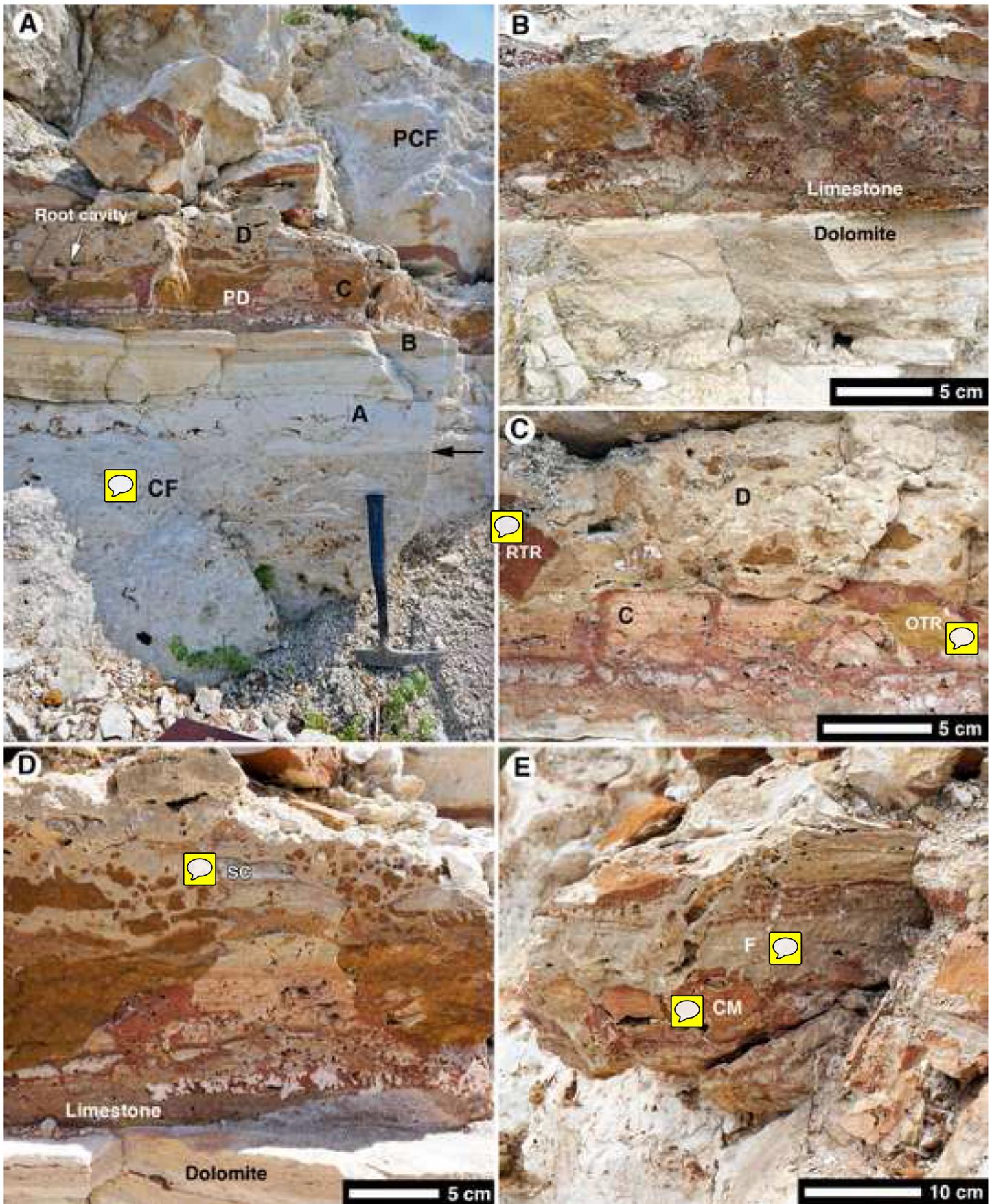


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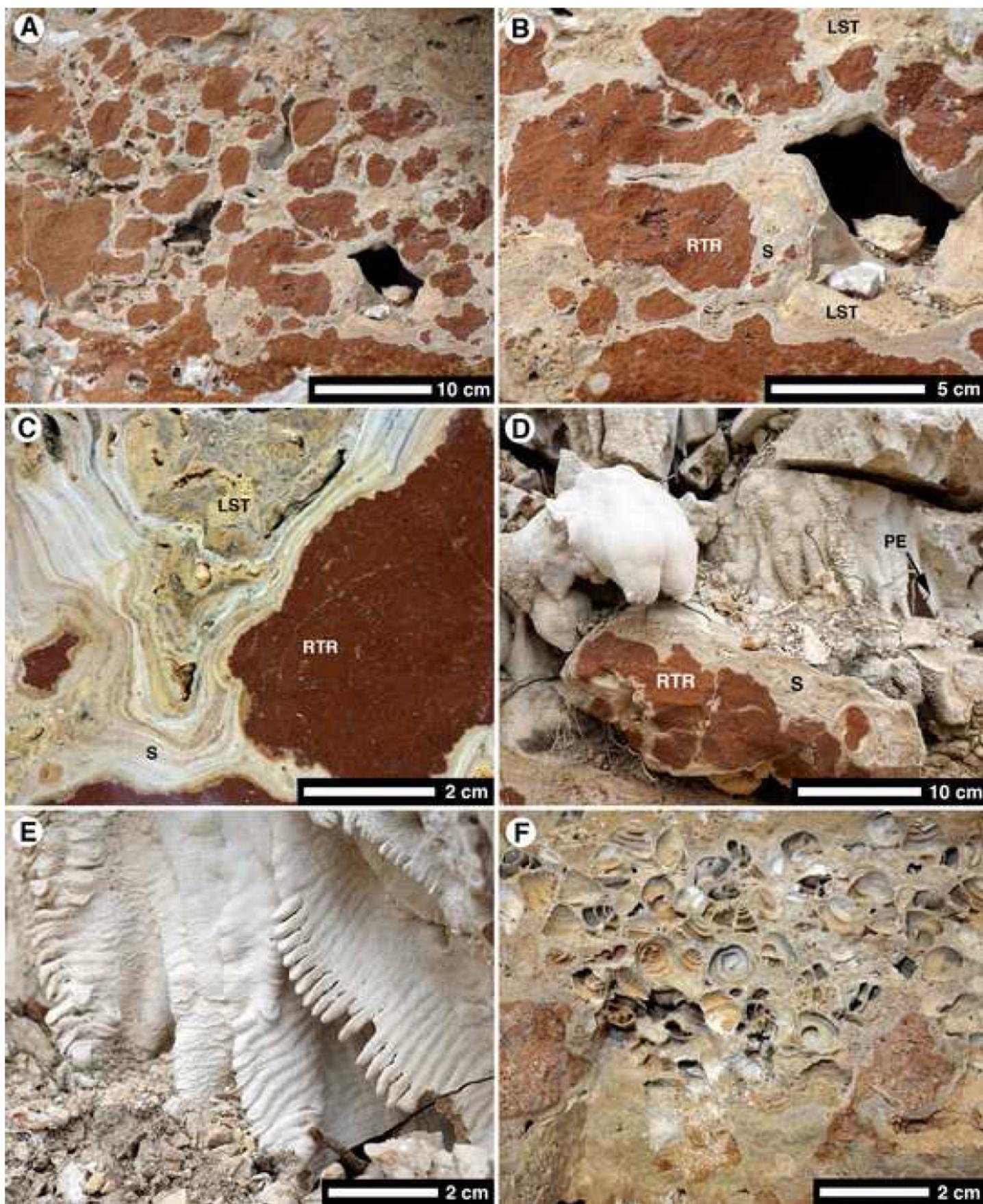


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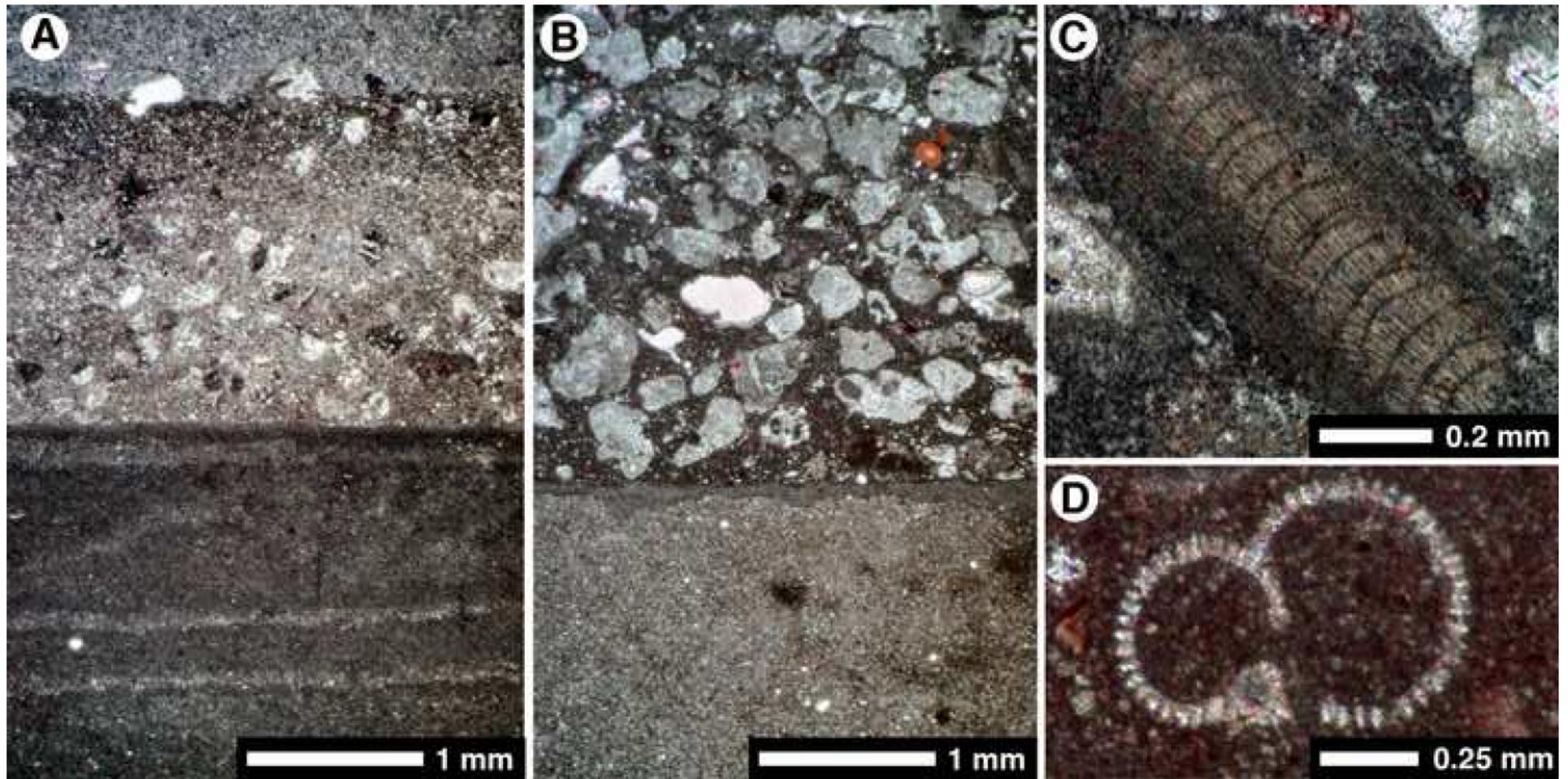


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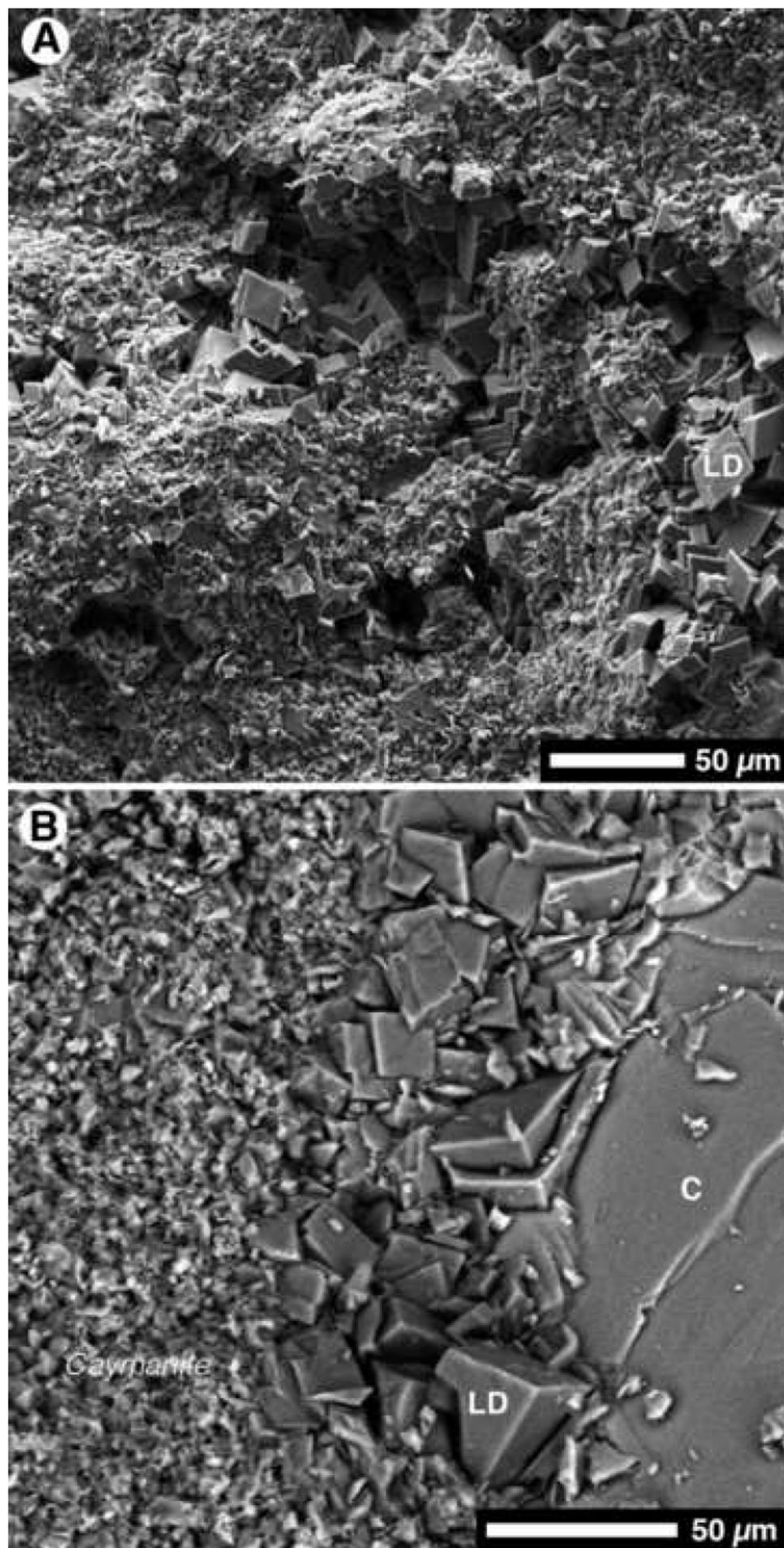


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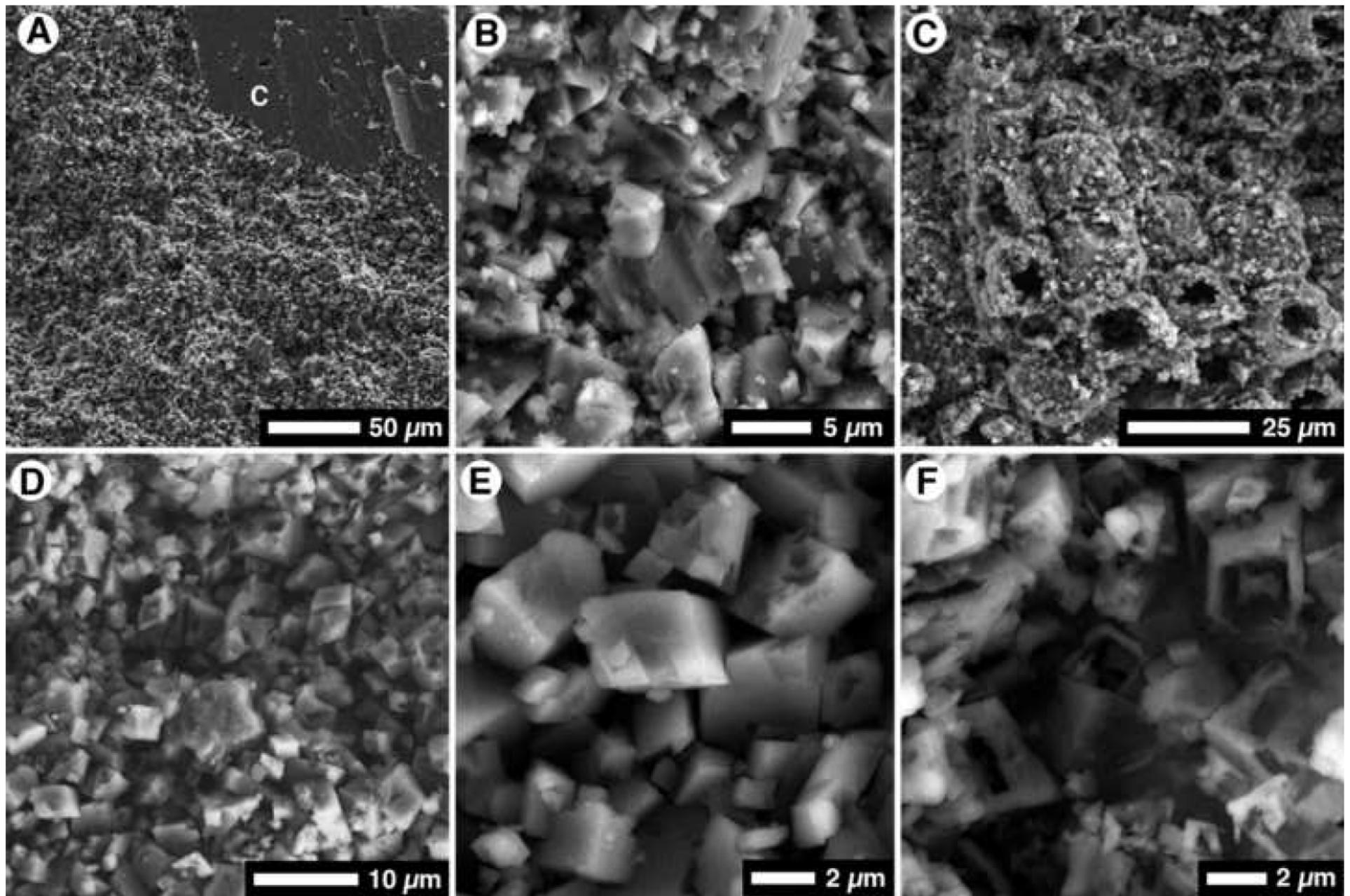


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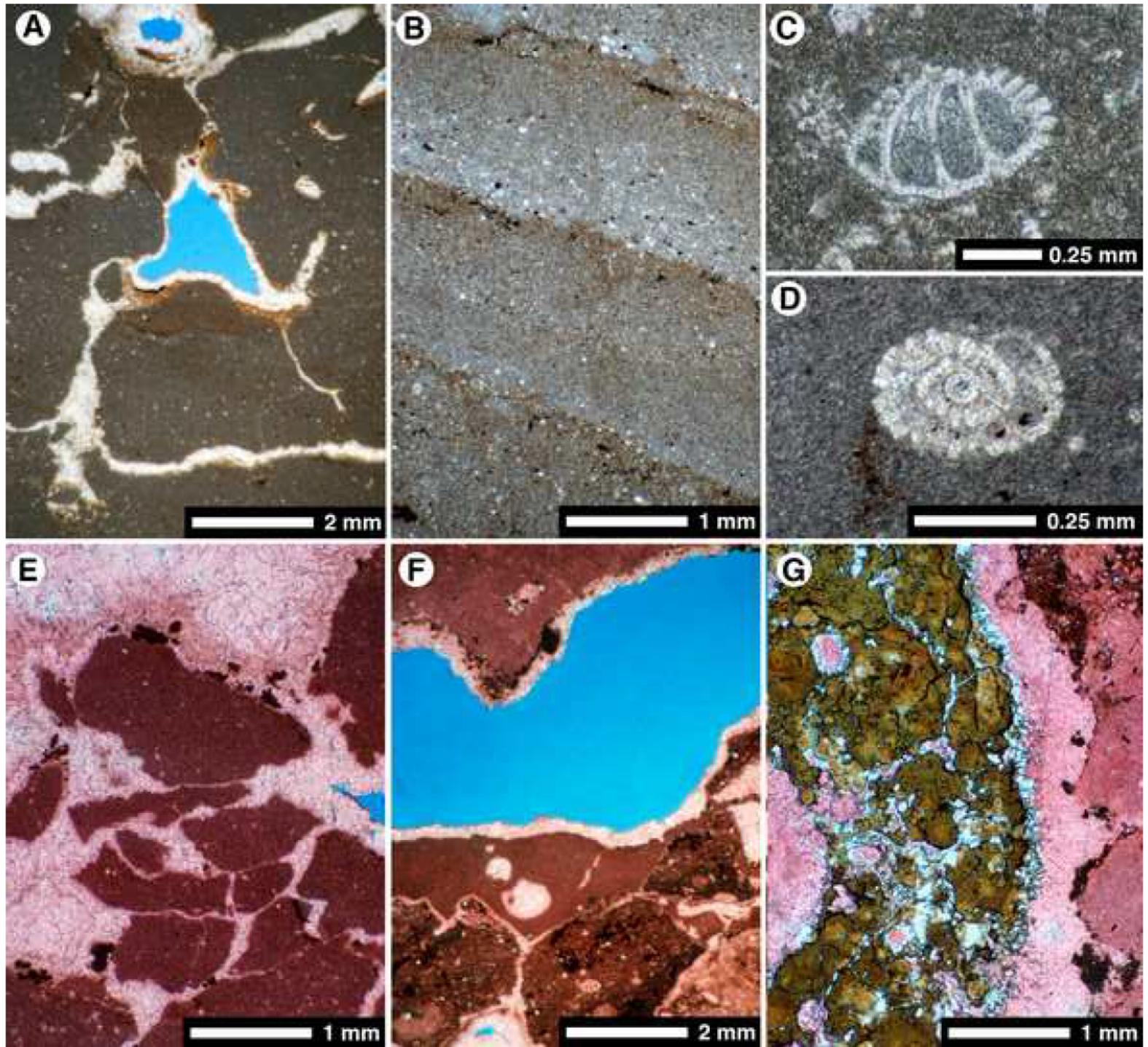


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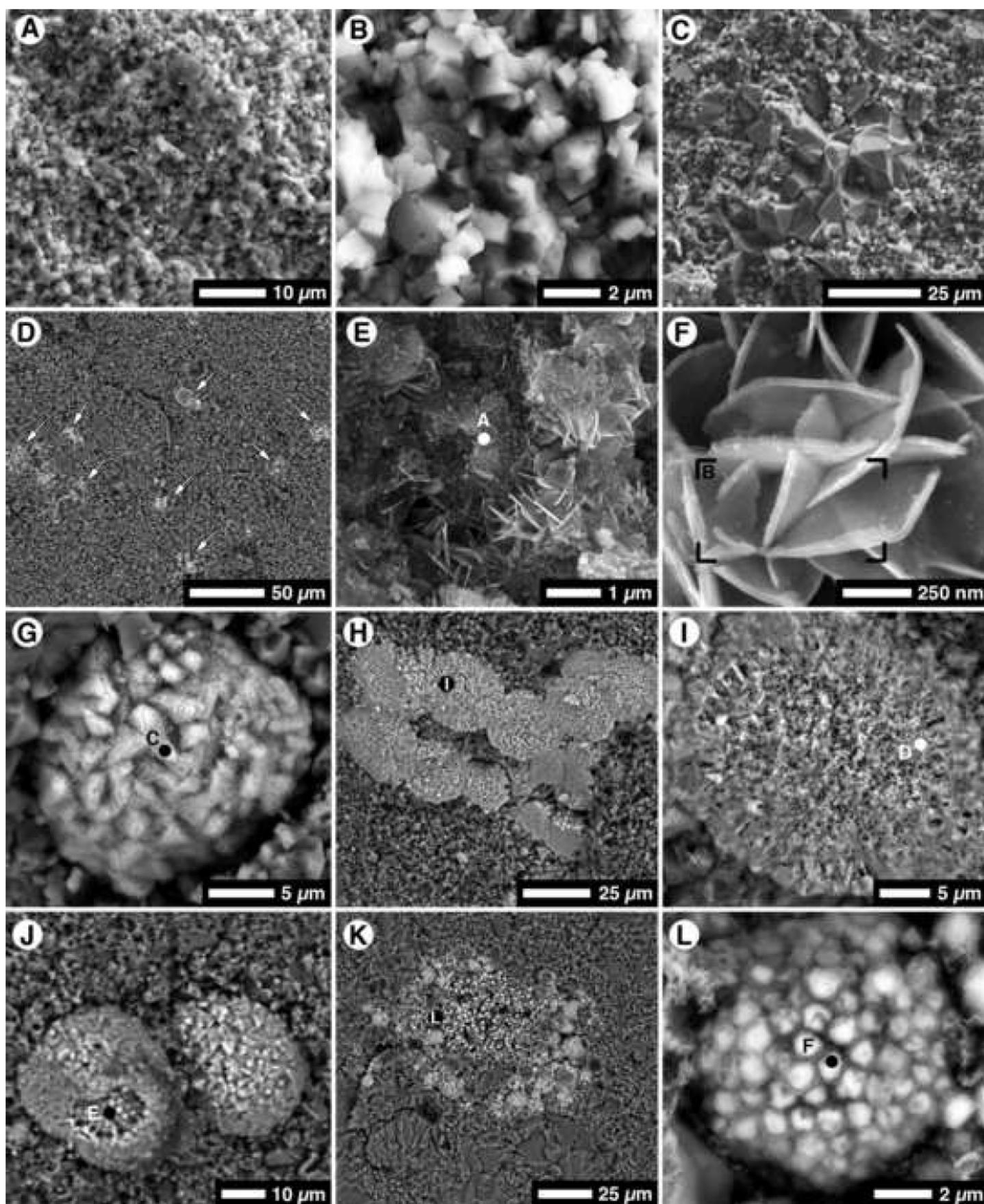


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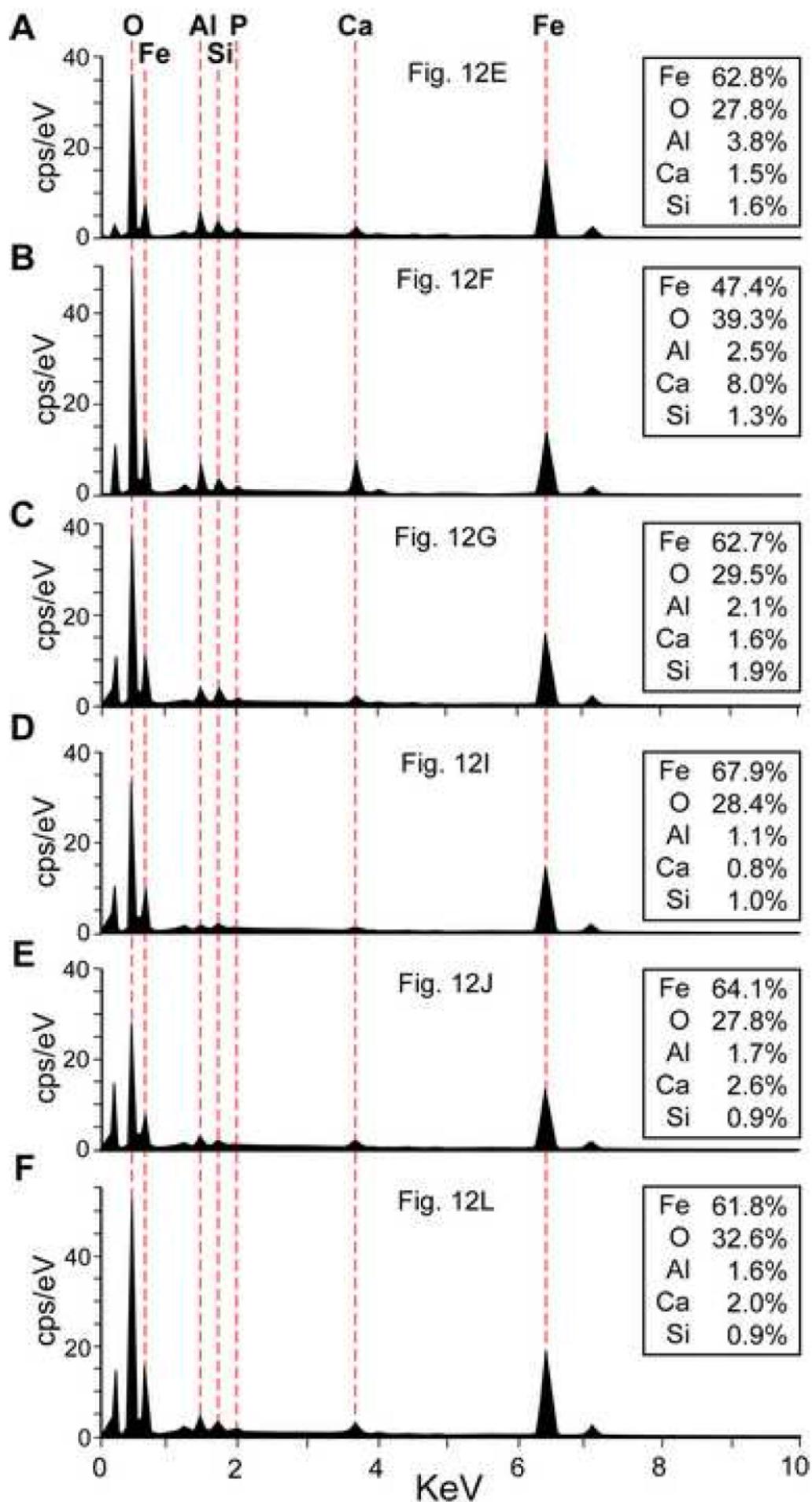


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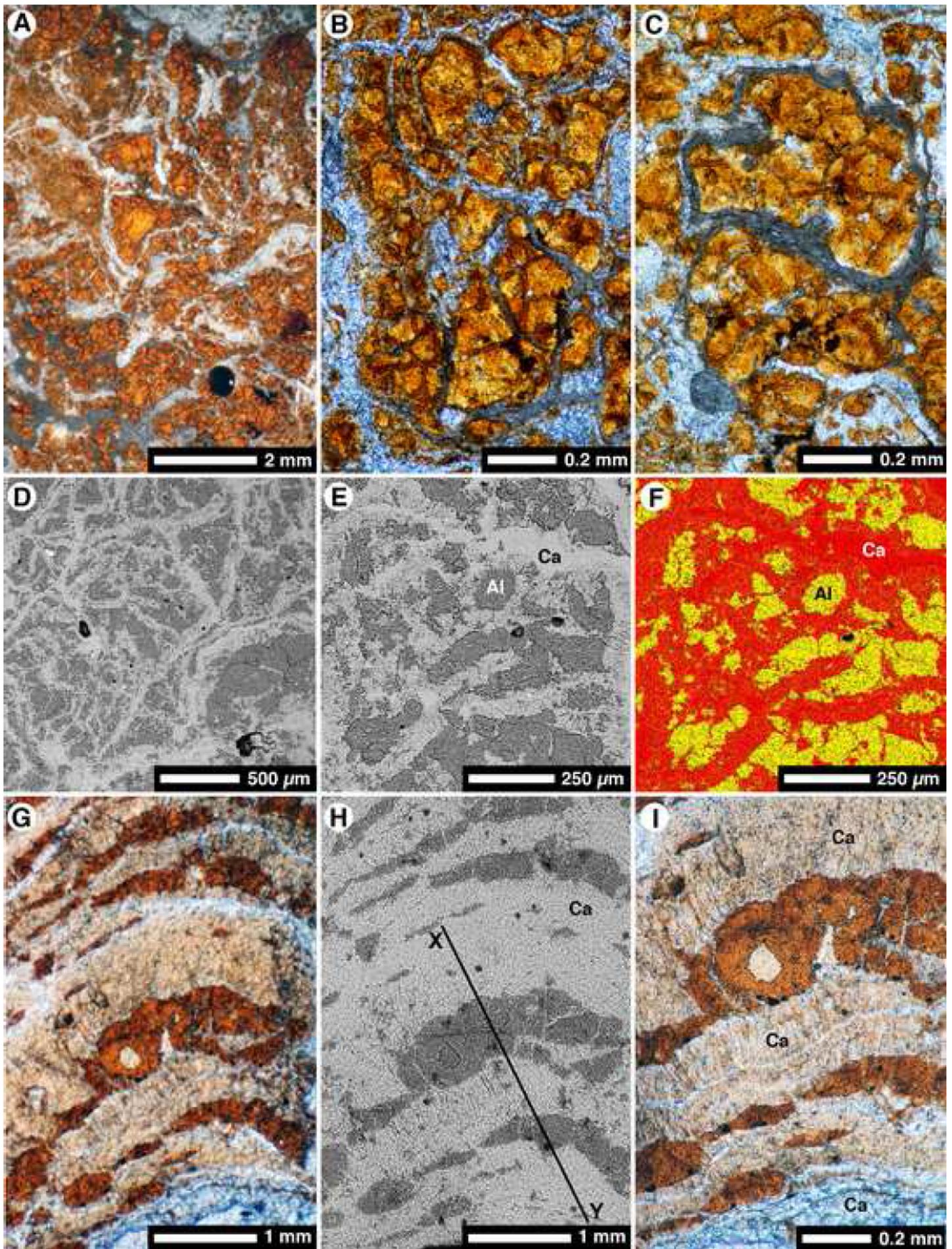


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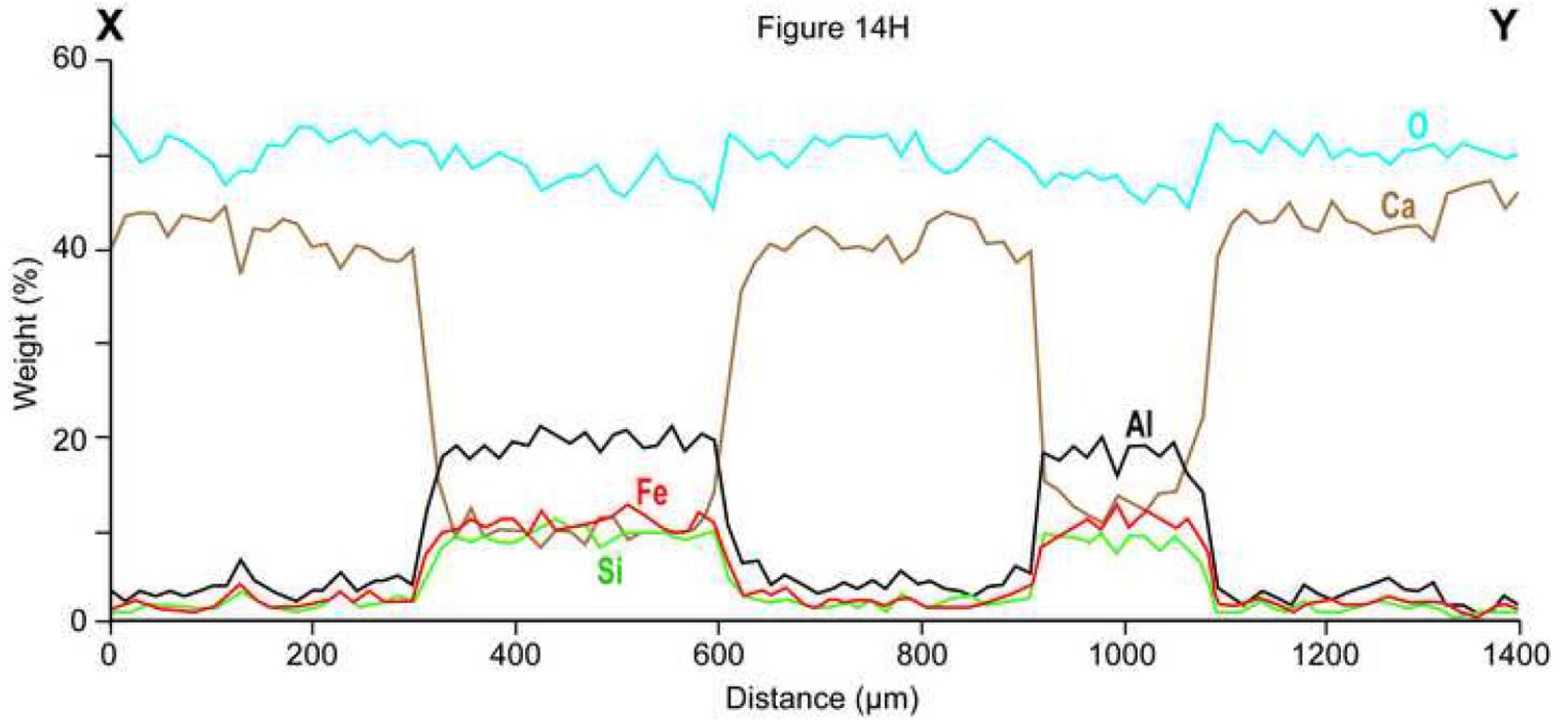


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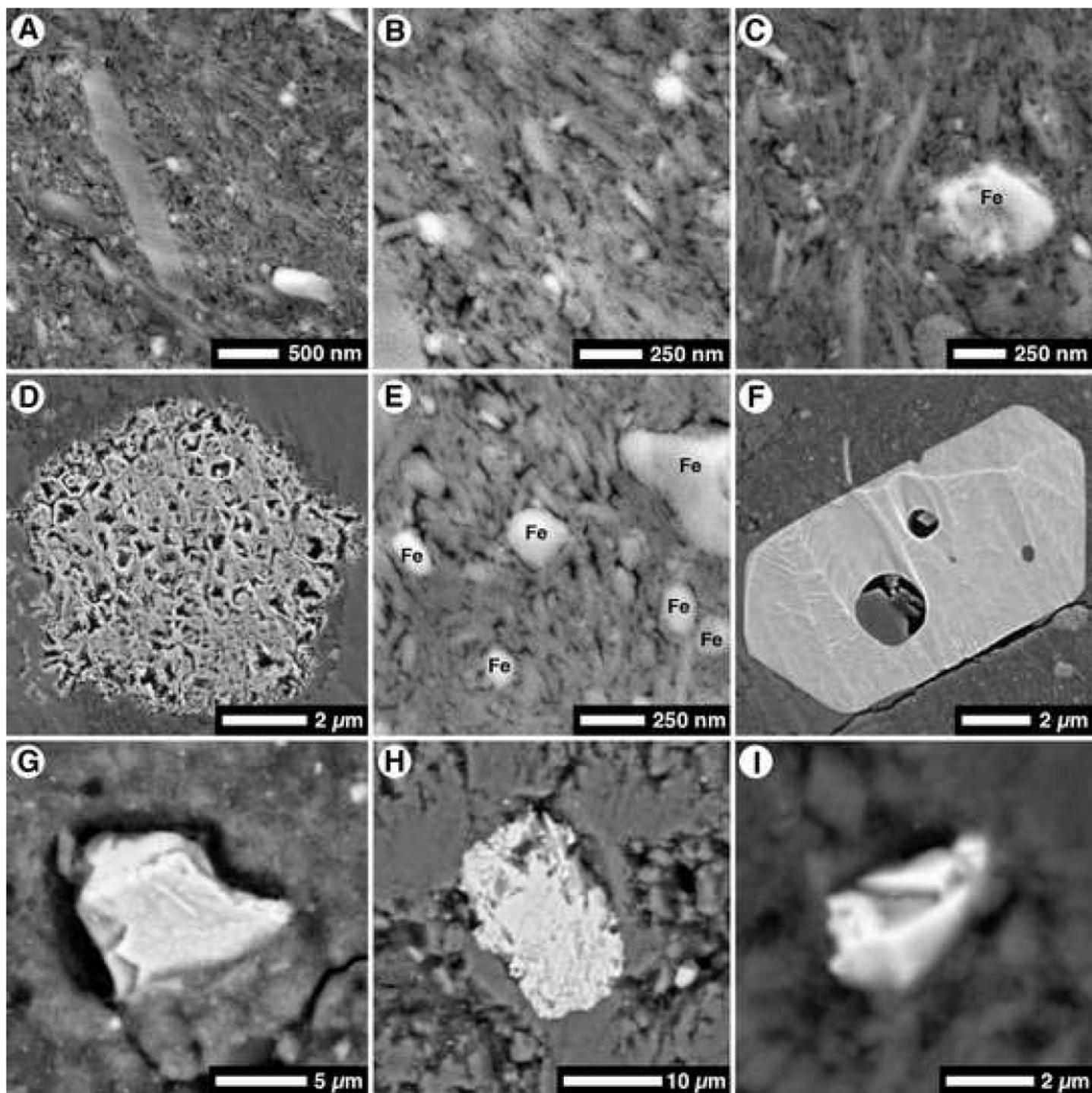


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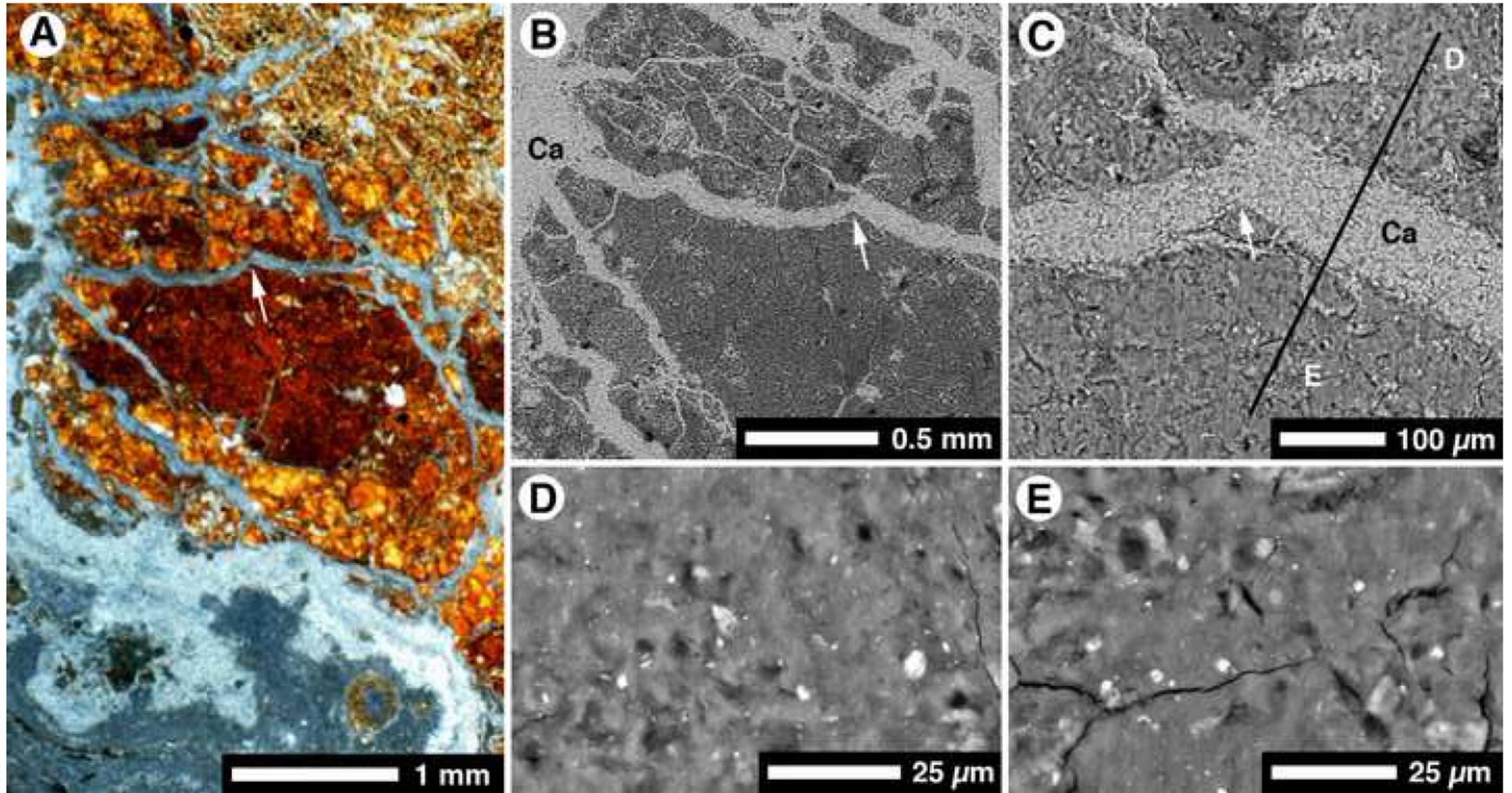


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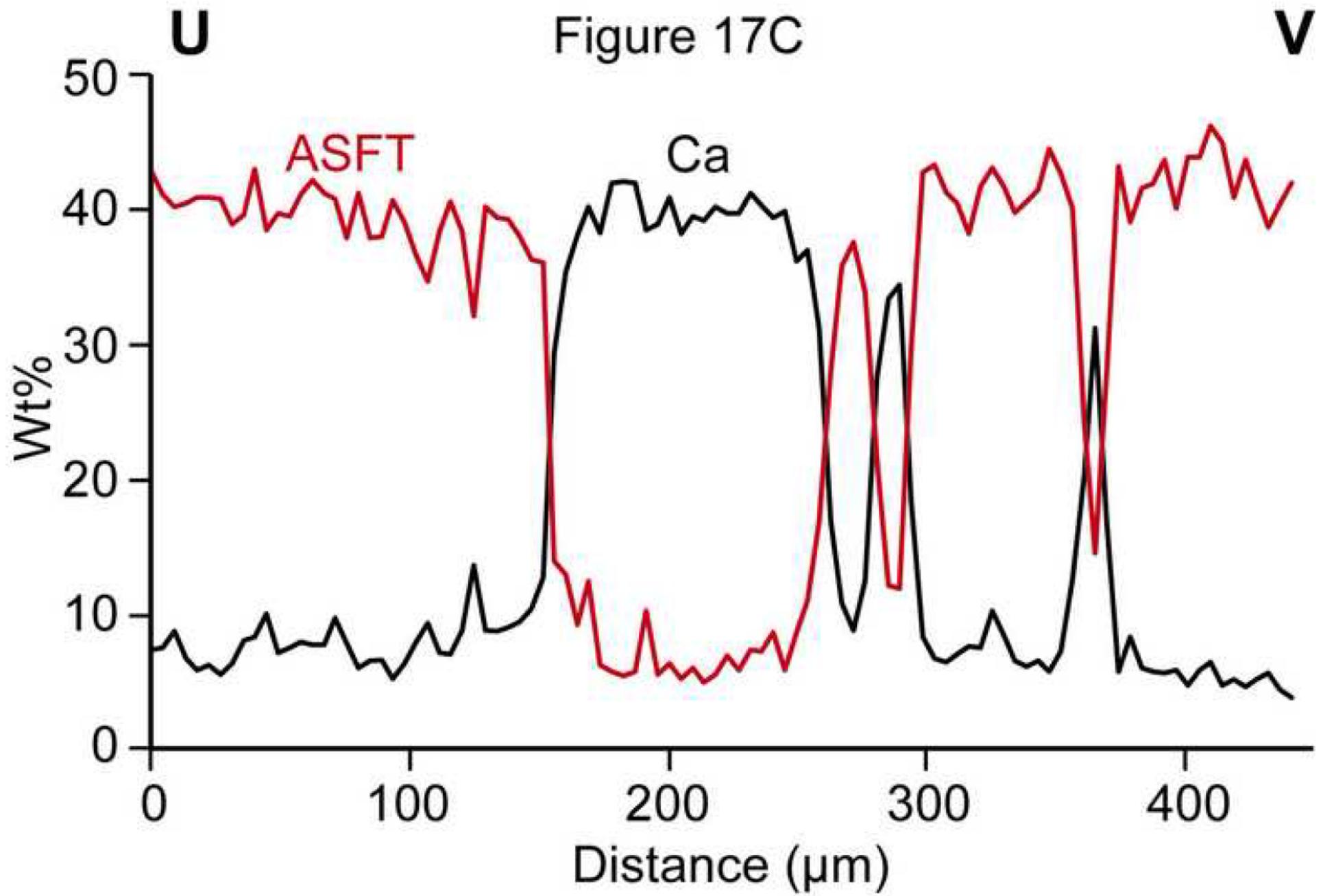


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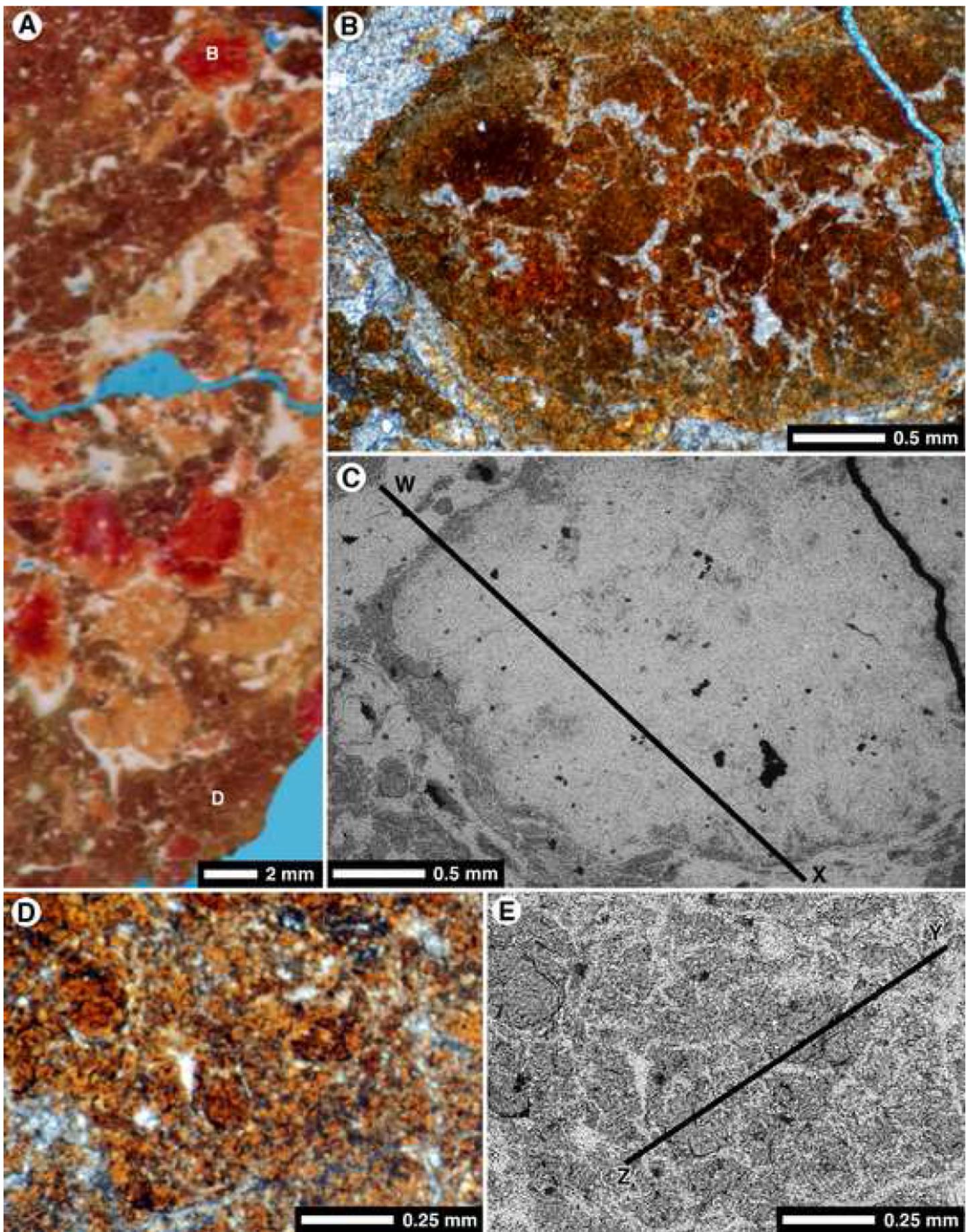


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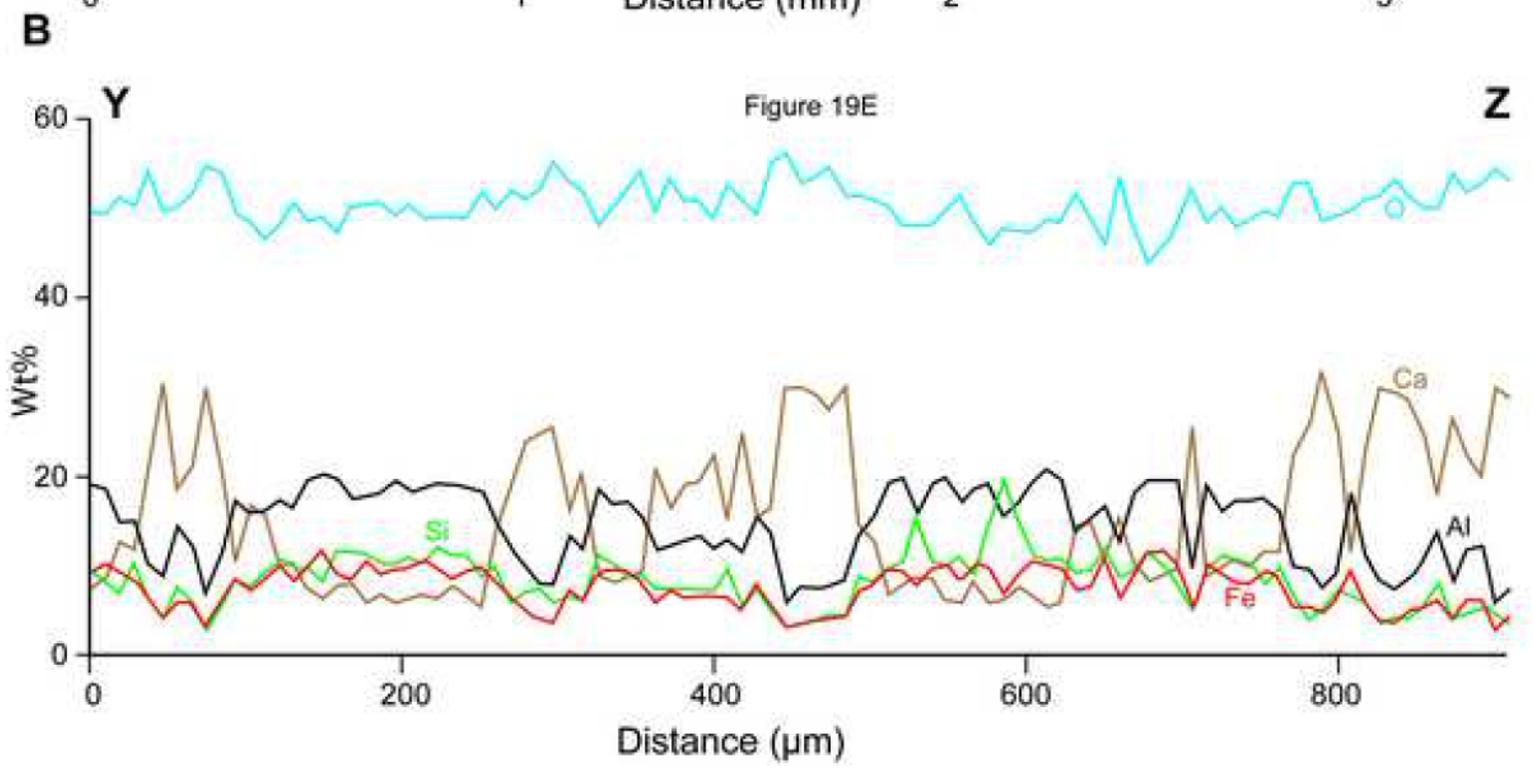
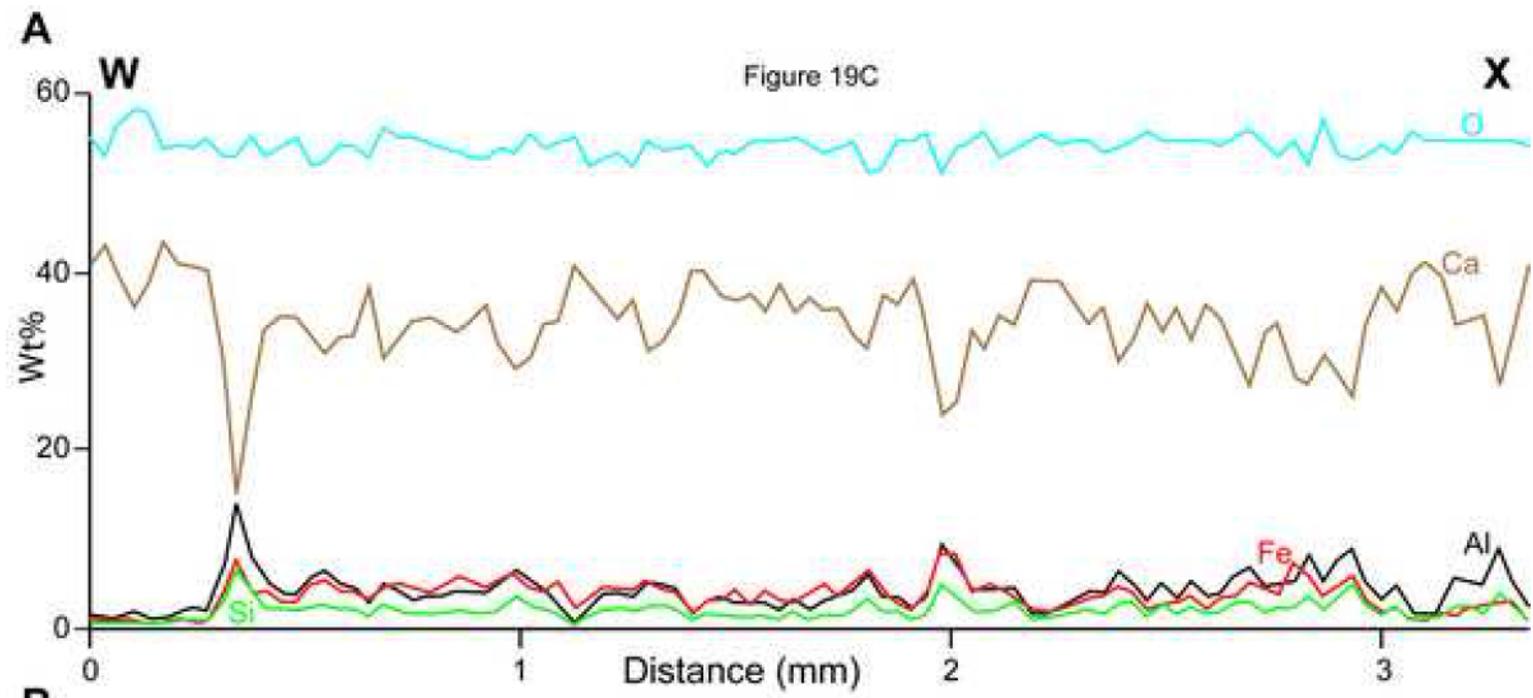


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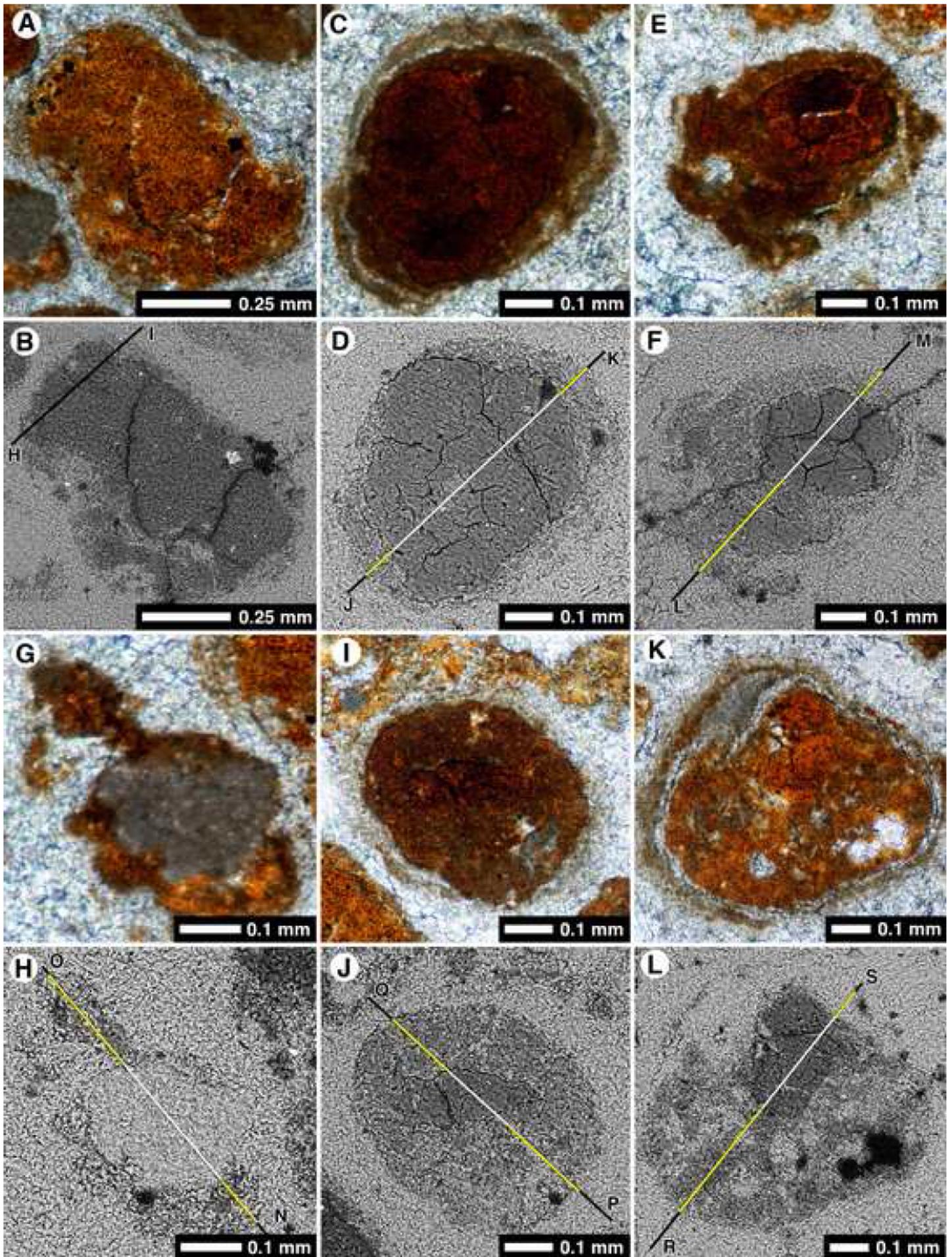


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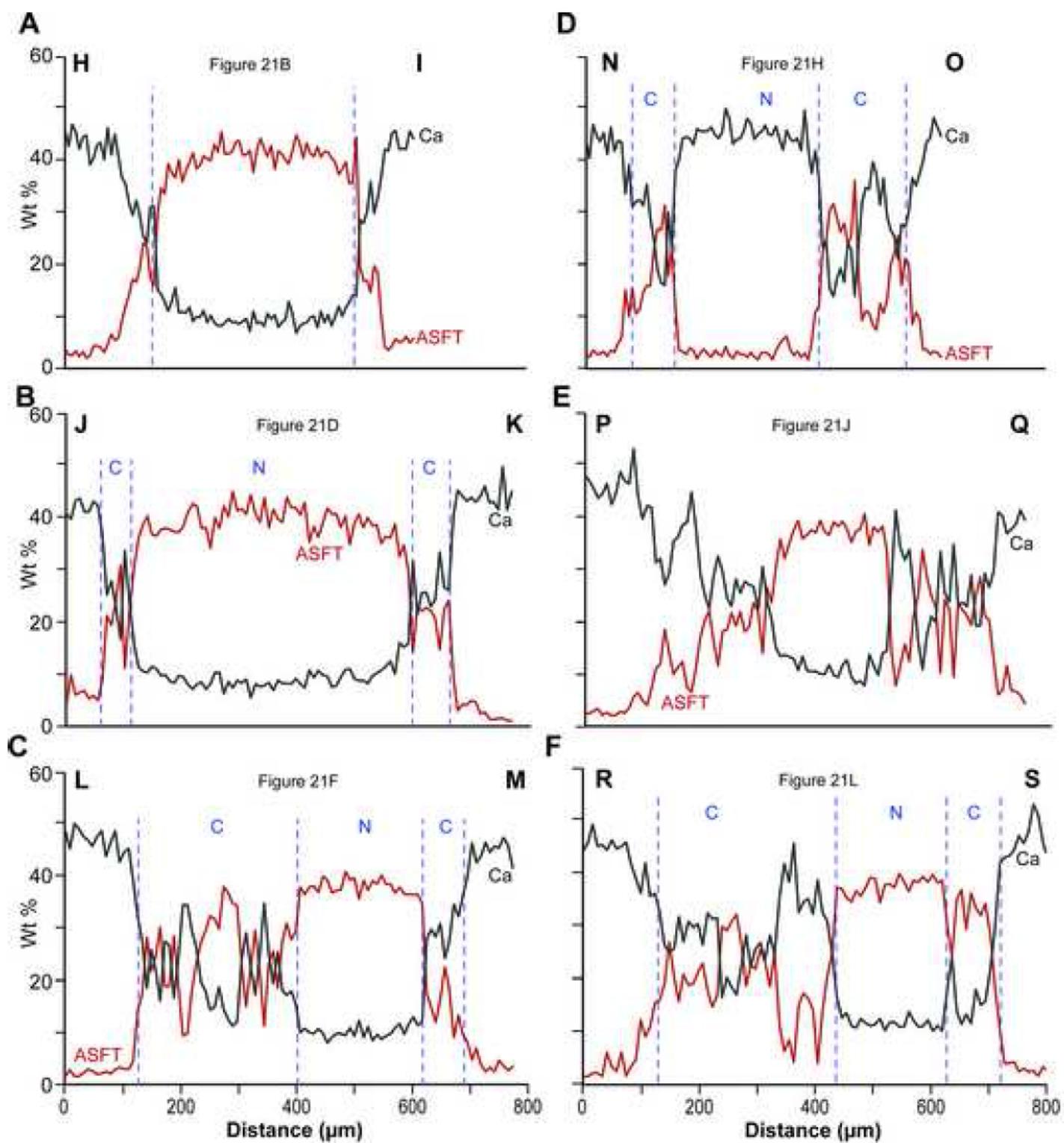


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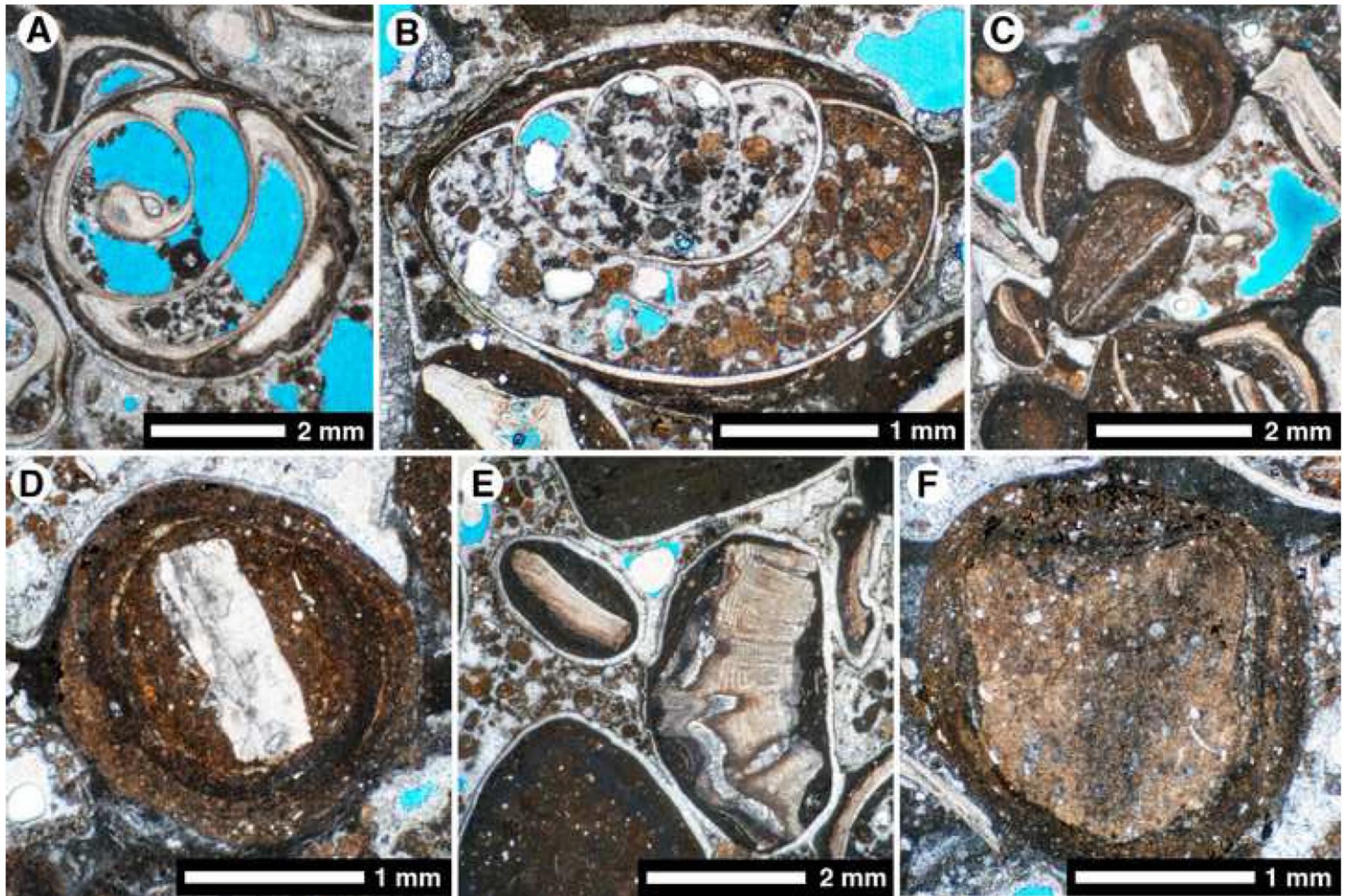


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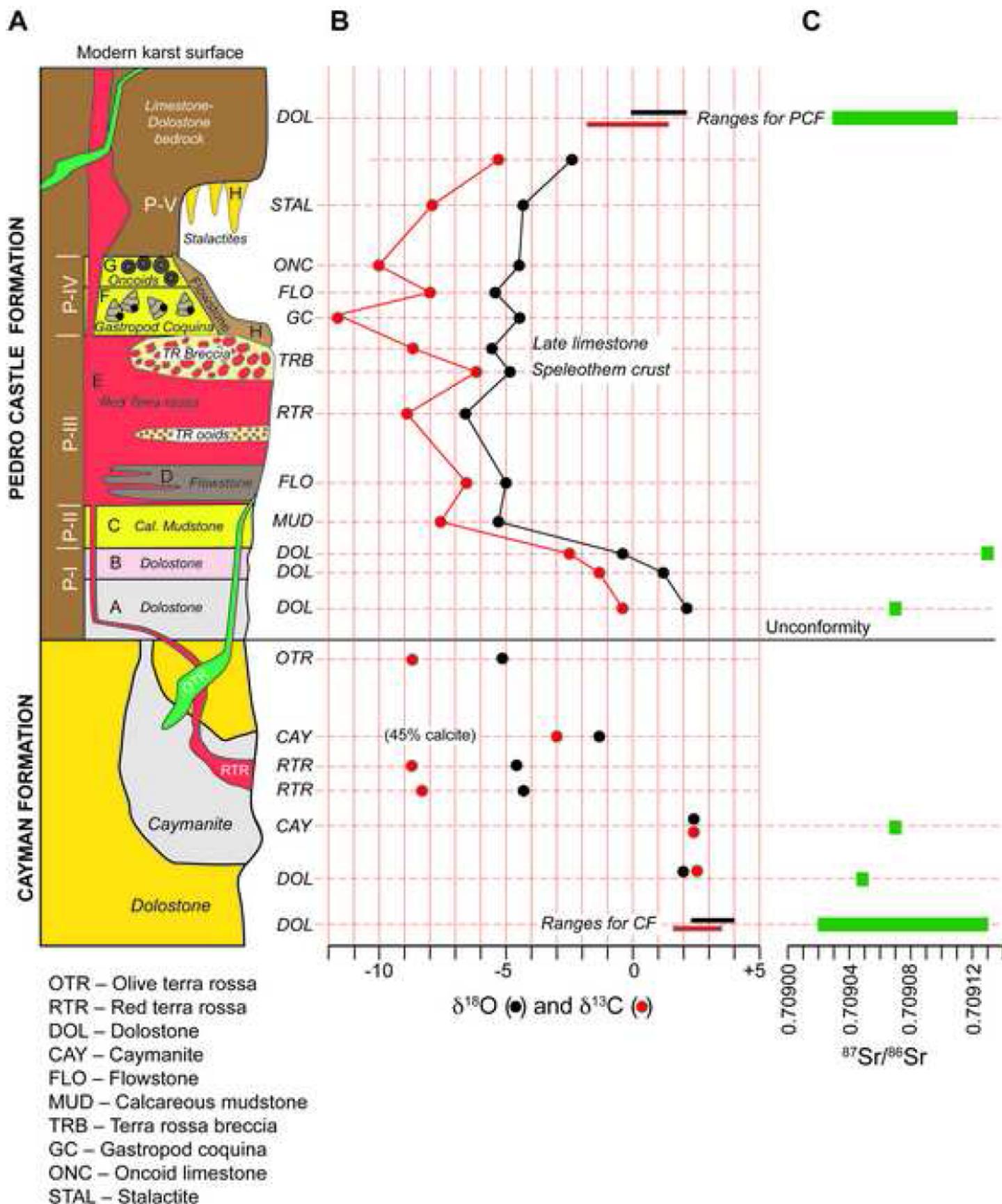


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