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1 **Testing whether early diagenesis of skeletal carbonate is different in non-**
2 **marine settings: contrasting styles of molluscan preservation in the Upper**
3 **Jurassic of Portugal**

4
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11
12 **Abstract**

13
14 This study tests the hypothesis that the early diagenesis of aragonite shells should differ
15 fundamentally between marine and freshwater environments. This is predicted to be the case
16 because aragonite is highly susceptible to dissolution in the TAZ (Taphonomically Active Zone) in low
17 energy marine settings due to acidity caused largely by the oxidation of H₂S generated by sulphate-
18 reducing bacteria, but reduced sulphide activity in freshwater settings should result in less early
19 dissolution of aragonite. To test this hypothesis a range of fresh-brackish-hypersaline and marine
20 limestones were sampled from the Upper Jurassic (mid Oxfordian) Cabaços Formation of central
21 western Portugal. In these freshwater and brackish deposits, molluscs are preserved mostly as
22 sparite shell replacements indicating that the original aragonite was preserved through the TAZ and
23 was later replaced during subsequent burial by calcite cement. In limestones deposited in more
24 marine to hypersaline settings, molluscan remains mostly consist of the calcitic layers of bimineralic
25 bivalves, as shell where the original was wholly calcitic, or as gastropod steinkerns. Exceptions occur
26 and reflect other factors such as higher energy conditions during deposition whereby organic
27 matter, as the drive for microbial decay processes, was removed. The implications for molluscan
28 preservation including some hydrocarbon reservoirs are discussed.

29 Key words: taphonomy; aragonite dissolution; non-marine; coquinas; sulphate reduction zone

30

31 **1. introduction**

32 The mechanisms and effects of the mobilization of carbonate during shallow burial in
33 marine waters have been appreciated for some time (e.g. reviews by Sanders, 2003; Cherns
34 et al., 2011): calcium carbonate, especially the more soluble aragonite, is dissolved in the
35 uppermost sediment layer largely as a result of acidity caused by the oxidation of H₂S, itself
36 produced by bacterial sulphate reduction. In lower energy settings organic matter can
37 accumulate in finer grained sediments, whether siliciclastic, carbonate or of mixed
38 composition, and sources the microbially mediated decay processes that then drive skeletal
39 carbonate dissolution, and re-precipitation.

40 While most of the effectively syndepositionally dissolved carbonate back-fluxes to the
41 water column, a fraction is re-precipitated as calcite in the sediment column in areas of
42 increased alkalinity, such as depths where sulphate reduction takes place, to produce
43 diagenetic bedding (Munnecke and Samtleben, 1996; Westphal and Munnecke, 2003;
44 Wheeley et al. 2008). This oxidized zone, effectively the taphonomically active zone (TAZ) of
45 Aller (1982) and Davies et al. (1989), will be controlled by diffusion from the overlying water
46 column, if oxygenated, and by mixing caused by bio-irrigation (mainly burrowing).

47

48 The hypothesis being tested in this study is that as oxidation of H₂S is a major factor
49 triggering dissolution of aragonitic shells, its effects should be less in many non-marine low-
50 sulphate settings such that aragonitic shells may have a greater likelihood of entering the
51 burial realm below the TAZ than in marine waters. This may seem a counter-intuitive
52 argument as many marine waters, especially tropical and sub-tropical ones are less likely

53 than freshwaters to be undersaturated with respect to aragonite. However, the
54 concentration of sulphate in freshwater ranges from ~10 to >500 μM , much lower than in
55 seawater (28 mM) (Holmer and Storkholm, 2001).

56 To test this hypothesis we have examined a range of fresh–brackish–marine and
57 hypersaline limestones from the Upper Jurassic Cabaços Formation of central western
58 Portugal (Figs 1A, 2). While the data set shows a general difference in the preservation of
59 former aragonite-bearing molluscan material between freshwater and other more saline
60 waters, other factors need to be considered such as rates of burial, organic-matter content
61 of the original sediment and the life position of aragonitic organisms relative to the TAZ.

62 **1.1. Molluscan shell taphonomy**

63 In low-energy marine settings, where organic matter can accumulate in the sediment
64 and trigger a range of microbial processes and other reactions, aragonite under-saturation
65 can lead to significant loss of shell material (e.g. Cherns and Wright, 2000; Cherns and
66 Wright, 2009; Wright et al., 2003). Many mollusc shells are aragonitic, some are bimineralic
67 (layers of calcite and aragonite) and some are entirely calcitic. The resulting fate of these
68 shells during passage through the TAZ depends on their mineralogy and on their residence
69 time in the zone of aragonite under-saturation (e.g. Sanders, 2003; Cherns et al., 2011).
70 Aragonitic shells are preferentially dissolved and the resulting moulds are largely destroyed
71 by bioturbation. Rarely steinkerns representing internal sediment fills of such shells can be
72 preserved. Complete micritisation of the aragonite shell leads to its replacement by calcite
73 but it is then difficult to distinguish such shell material from other forms of micritised,
74 originally calcitic bioclasts. If the aragonite bypasses the TAZ into the final burial zone (FBZ;
75 Olszewski, 2004), such as in the taphonomic windows identified by Cherns et al. (2008),

76 aragonite can persist or be congruently dissolved leaving a mould which can remain as a
77 pore, or be filled by calcite cement, or be incongruently replaced (neomorphosed) to retain
78 some palimpsest shell structure. Bimineralic shells retain their thin outer calcitic layer but
79 the aragonitic layer(s) undergoes the same possible pathways as for wholly aragonitic shells.
80 The remnant calcitic shell layer is commonly very thin and resembles the filaments
81 described from various marine successions. Wholly calcitic mollusc shells generally survive
82 little altered mineralogically into the FBZ.

83

84 **2. Cabaços Formation**

85

86 The Cabaços Formation (mid-Oxfordian; e.g. Ramalho, 1981; Azerêdo et al. 2002a,b;
87 Kullberg and Rocha, 2014) crops out extensively in the Mesozoic Lusitanian Basin in west
88 central Portugal (Figs 1, 2) (e.g. Wilson, 1979; Leinfelder and Wilson, 1998; Azerêdo et al.,
89 2002a,b). It typically ranges from ca. 40-150 m in thickness at outcrop (but locally
90 subsurface data point to 180-200 m or more) and disconformably overlies marine Middle
91 Jurassic units. The three most representative of these are the mid-outer ramp Cabo
92 Mondego formation (an informal unit) sensu Azerêdo et al. (2003), corresponding to the
93 distal marine facies associations broadly named "Brenha" in industry reports, and the inner
94 ramp Santo António-Candeeiros and Serra de Aire formations (Azerêdo, 2007),
95 corresponding to the proximal marine facies associations broadly named "Candeeiros" in
96 industry reports (Fig.2).

97 The basal Cabaços typically consists of medium to thick-bedded non-marine limestones,
98 marls and lignites with a broad thickening trend towards the west and south from 12–16 m
99 at Serra dos Candeeiros, Maciço Calcário Estremenho (Fig. 1B), where they are underlain by

100 c. 8-14m of pedogenic/black-pebble limestones and ferruginous marls (Azerêdo et al. 2002a,
101 b), to over 20m at Pedrógão, to over 40 m at Serra d'El-Rei (Fig. 1B). In general it is typified
102 by intercalations of marls, marly limestones, ferruginous limestones/marls and, locally,
103 lignites and lignitic clays/marls, black-pebble limestones, and less commonly with pedogenic
104 limestones (at the base), and isolated levels of microbial laminated micrites; deltaic
105 sandstones and coral-oyster bioherms occur very locally, as at Cabo Mondego (Wright,
106 1985; Azerêdo et al. 2002b). Desiccation polygons up to 0.3 m in diameter are present
107 locally and ferruginized irregular surfaces cap some beds such as at Pedrógão. In addition
108 irregular fenestrae and vadose cements occur but are not widespread. Poorly preserved
109 porostromate tubes are found in mm–cm grey calcimudstones to floatstones and at some
110 localities, the lumps constitute small to large (several centimetres) oncoids.
111 The palaeobiota is typified by non-marine taxa, in particular highly abundant charophytes
112 and ostracods, common gastropods, bivalves, fossil wood remains, diverse palynomorphs of
113 dominant continental source and rare reptile teeth. Extensive bioturbation has occurred at
114 some levels.

115

116 The upper part of the Cabaços Formation typically consists of thin-bedded (locally
117 medium-bedded), heterogeneous non-cyclic limestones, marly limestones, microbial
118 laminites at places bituminous, and evaporites (Azerêdo et al., 2002b). Within the basin the
119 total thicknesses in outcrop are extremely variable from as little as 15 m at Vale de Ventos,
120 to at least 30 m at Pedrógão, 60m at Cabo Mondego but, in the Benfeito-1 well (Montejunto
121 region), it reaches over 100 m (Fig. 1B). Thus, it is difficult to pick out a clear lateral
122 thickening/thinning trend. In general the limestones consist of bioclastic calcimudstones to
123 packstones, with minor bioclastic grainstones which can be indistinctly to clearly graded,

124 with parallel and low-angle cross-lamination and may show erosive basal surfaces
125 developed on the underlying micrite-dominated levels. Rare oolitic-intraclastic grainstones
126 also occur. Desiccation cracks are common and dinosaur trackways also occur. Ostracods,
127 including as coquinas, are abundant as well as charophyte gyrogonites and less common
128 stems. Gastropods and bivalves are very common, associated with dasyclads (at places
129 abundant), rare agglutinate-walled foraminifers, serpulids, very rare echinoid remains and
130 lower-diversity palynomorphs. Bioturbation effects are common. Gypsum crystal
131 pseudomorphs and nodules are common, typically calcitised or silicified, and rare metre-
132 scale evaporite breccias occur. Metre-thick beds of anhydrite are recorded in the subsurface
133 (namely, at Benfeito-1 well). The organic-rich deposits of the Cabaços Fm., corresponding to
134 different lithologies (lignites, lignitic clays/marls, bituminous marls/marly limestones and
135 bituminous microbial laminites), have yielded heterogeneous values for geochemical
136 parameters, namely TOC, across the basin. For instance, Silva et al. (2014) record TOC values
137 ranging 0.26–4.67 wt% at Pedrógão, 0.33–30.56 wt% at Cabo Mondego (material heavily
138 impregnated with hydrocarbons) and 0.43–11.64 wt% at Vale de Ventos, whereas Spigolon
139 et al. (2011) indicate 2.87–4.93 wt% (outcrop samples) and Gonçalves et al. (2015) <1 wt%
140 (borehole samples) at different places in the Montejunto region. The palynofacies is also
141 variable but overall data show that the particulate organic matter is mostly of continental
142 origin, punctuated by minor events of marine-influence, which increases towards the upper
143 part of the unit (Barron and Azerêdo, 2003; Spigolon et al., 2011; Silva et al., 2014;
144 Gonçalves et al., 2015).

145

146 This study focuses on the Pedrógão section (Fig. 1B), which is one of the most
147 representative and well documented outcrops of the Cabaços Formation, for which a
148 detailed palaeontological framework is already available.

149

150 2.1. Pedrógão Section

151

152 The well exposed coastal section at Pedrógão beach (Fig. 1B) exhibits an Upper Callovian-
153 Oxfordian succession that has been described in detail by Azerêdo et al. (2002a, b) and
154 further addressed in several thematic works on microfossil groups, namely ostracods (Cabral
155 et al., 1998; Azerêdo et al., 2002a; Pais et al., 2016), charophytes (Grambast-Fessard and
156 Ramalho, 1985; Pereira et al., 1998, 2003; Azerêdo et al., 2002a) and palynomorphs (Barron
157 et al., 1999; Barron and Azerêdo, 2003). Azerêdo and Cabral (2004), using a range of
158 palaeontological (chiefly the ostracod associations) and sedimentological data, identified in
159 the Oxfordian deposits four facies/palaeoecological subdivisions, which are here labelled
160 Units 1-4, updated and taken as independent background to test the facies interpretations
161 of the present study (Fig. 3).

162

163 Note however, that ostracod assemblages were generally recovered from marls which
164 allow the removal of whole carapaces, whereas most mollusc samples allowing
165 determination of shell preservation were taken from indurated limestones, thus a direct
166 comparison between these sample sets is not possible. The emphasis is therefore placed on
167 trends in terms of salinity and preservation rather than identifying the exact conditions for
168 specific horizons. At Pedrógão the more thickly bedded lower unit (Unit 1) is overlain by thin

169 bedded upper Cabaços which can be sub-divided into three sub-units (Units 2-4; Azerêdo
170 and Cabral, 2004).

171

172 2.1.1. Unit 1

173

174 The lowermost Cabaços, 24 m thick was designated as Unit 1 by Azerêdo and Cabral
175 (2004) (Fig. 3), and is characterised at outcrop by mainly matrix-dominated limestones, with
176 lignites and lignitic marls. Evidence of exposure is not common and includes desiccation
177 cracks at three levels, rare fenestrae and possible evidence of pedogenesis at one bedding
178 surface.

179

180 The fauna contains a range of mainly freshwater to slightly mesohaline ostracods,
181 documented in detail by Azerêdo and Cabral (2004) such as the genera: *Theriosynoecum*,
182 *Darwinula/Alicenula*, *Mantelliana*, *Sinuocythere*, *Timiriasevia* and *Klieana*. The ostracod
183 fauna generally increases in diversity towards the top of this unit, almost always with the
184 genus *Theriosynoecum* dominant, but there are clear variations in the dominant species (of
185 *Theriosynoecum*, *Darwinula*, rarely *Mantelliana* or *Sinuocythere*), as well as ornamentation
186 styles and abundance. The most common are: *Theriosynoecum* spp., indicating freshwater
187 to slightly mesohaline conditions (Carbonel et al., 1988), found in assemblages as the
188 dominant species, typically very abundant, exhibiting ornamentation variations;
189 *Darwinula/Alicenula* spp., occur also in assemblages with one species dominant and very
190 abundant, as very large forms, indicating slightly more salinity-tolerance than
191 *Theriosynoecum*, but still freshwater to weakly mesohaline conditions as *Darwinula* can

192 occur today at salinities of possibly over 10 ‰ (Carbonel et al., 1988). The living species *D.*
193 *stevensoni* (Bradley and Robertson) tolerates an increase in salinity up to a maximum of
194 15‰ (Hiller, 1972, in Meisch, 2000). *Mantelliana* spp., becomes more common upwards in
195 the unit, indicating fresh?/brackish, meso- to polyhaline conditions, possibly tolerating
196 salinities up to 30‰ (Carbonel et al., 1988). Starting from 14.55m above the base of the
197 section, there are a few levels of small-sized, smooth or almost smooth specimens of the
198 euryhaline *Sinuocythere pedrogaensis* Cabral and Colin (Colin et al., 2000; Azerêdo and
199 Cabral, 2004) (*cf.* palaeocology in Colin et al., 2000). In addition *Timiriasevia* and *Klieana*,
200 both mainly oligohaline forms (Carbonel et al., 1988) are present at only a few levels.

201

202 An abundant and diverse charophyte flora is present (Grambast-Fessard and Ramalho,
203 1985; Pereira et al., 1998, 2003; Azerêdo et al., 2002a, b), including horizons of *in situ*
204 calcite-encrusted stems indicating former presence of *Chara* meadows (Fig. 4A). There is a
205 diverse palynomorph assemblage (namely *Corollina cf. torosus* and spores related to the
206 Bennettitales and Cycadales), together with the typically freshwater alga *Botryococcus*, with
207 minor marine-influenced contributions (Barron et al., 1999; Barron and Azerêdo, 2003). The
208 palynofacies is mostly of a terrestrial origin with sporadic marine influence with phytoclasts
209 and amorphous organic matter (AOM) increasing/decreasing oppositely as the result of
210 periods of lower/higher environmental restrictions and preservation conditions (Silva et al.,
211 2014). Remains of fully marine forms are very rare.

212

213 Former aragonitic molluscs, mainly gastropods (locally abundant small-sized forms), are
214 preserved predominantly as sparite shell replacements lacking any remains of wall structure
215 (Fig. 4B). Bimineralic shells show a well-preserved outer, thin calcitic layer with a sparitic

216 former aragonitic layer (Fig. 4C). Thin (0.1mm) calcitic laminae of bimineralic forms (here
217 referred to as filaments) also occur at several levels but are not abundant and are typically
218 highly comminuted.

219

220 In summary, this lowermost unit in the Cabaços of Pedrógão has been interpreted as a
221 mainly freshwater deposit, representing more or less permanent shallow coastal lakes
222 (Azerêdo et al., 2002b), with vegetated fringes and/or marshes. The ostracod fauna
223 generally increases in diversity towards the top of this unit, but almost always with the
224 genus *Theriosynoecum* dominant, but the more common presence of *Mantelliana* and of
225 *Darwinula* suggesting a shift towards slightly more salinity-tolerant populations. The
226 appearance of *Sinuocythere pedrogaensis* is further evidence for episodes of slightly higher
227 salinity.

228

229 Preservation of former aragonitic mollusc shells as sparite replacements indicates that
230 the original aragonite survived through the TAZ and was later replaced during subsequent
231 burial and congruent dissolution by calcite cement. Bimineralic shells that show a well-
232 preserved outer, originally calcitic layer and the sparitic former aragonitic layer(s) (Fig. 4C)
233 are also evidence that aragonite survived the TAZ. These bivalves tolerate oligohaline but
234 not freshwater conditions (Fürsich 1981; Fürsich et al. 2009). The presence of some filament
235 shell laminae, here interpreted as calcitic layers of bimineralic bivalves left after dissolution
236 of aragonite laminae implies that some early dissolution of aragonite is likely to have
237 occurred locally. The filament material is highly comminuted and would have been readily
238 transportable and could be allochthonous. It is noteworthy that the ostracods from marls
239 immediately below or above limestones with higher proportion of calcitic filaments also

240 show small-scale variations in faunal composition pointing to slight salinity increase (within
241 the 'larger-scale' freshwater/brackish assemblages). Although this may be circumstantial
242 evidence, it supports the possible occurrence of very short flooding episodes, bringing more
243 saline water and with it transported, reworked filament material.

244

245 2.1.2. Unit 2

246

247 The nearly 8 m thick Unit 2 (Fig. 5) has predominantly matrix-rich limestones with some
248 grainstones. Microbial laminites occur, and desiccation cracks are present at eight levels,
249 some associated with fenestrae. Nodular textures occur at some levels forming
250 conglomerate-like bed surfaces and irregular micro-cracking which resemble pedogenic
251 alteration as described from the Cabaços Formation at Vale de Ventos, Serra dos Candeeiros
252 (Azerêdo et al., 2002b), (Fig. 1B). Ostracod faunas, in contrast to previous unit, are
253 dominated by the euryhaline species *Sinuocythere pedrogaensis*, as small specimens,
254 smooth but also slightly reticulated, suggesting some ecological change, although the mainly
255 oligohaline *Theriosynoecum* and some other genera present in Unit 1 also occur. A new
256 species belonging to a genus close to *Klieana* (*Klieana?* sp.) appears, probably more
257 euryhaline, due to its morphological characteristics, as suggested by the almost complete
258 absence of median vertical sulcus; overall, the assemblage suggests brackish to slightly more
259 saline affinities.

260

261 Charophyte gyrogonites while ubiquitous are less abundant, stems are rare, and thin
262 horizons packed with stems of the dasyclad *Barattoloporellopsis lusitanica* (Ramalho),
263 formerly *Heteroporella lusitanica* (Ramalho) (see Granier et al., 2017) occur. Benthic

264 foraminifera such as *Kurnubia palastiniensis* Henson and other agglutinate-walled forms
265 occur at some levels (Azerêdo et al., 2002a; Azerêdo and Cabral 2004). The palynofacies
266 shows assemblages of continental and less common marine contributions, namely
267 extremely rare freshwater palynomorphs, *Botryococcus* and dinoflagellate cysts, and high
268 amounts of AOM (Barron et al., 1999; Barron and Azerêdo, 2003; Silva et al., 2014).

269

270 Thick shelled *Isognomon rugosus* oysters (Fursich et al. 2009) form shell concentrations
271 associated with serpulid buildups (Fig.5A, B). The most common fossils are calcitic shells of
272 *Praeexogyra pustulosa* oysters in shell pavements, and there are horizons with filaments of
273 bimineralic bivalves. Another shell horizon above a desiccation cracked surface has
274 concentrations of neomorphosed shells of shallow burrowing nuculoid bivalves and small
275 gastropods. Fursich et al. (2009) and Fursich (1981) interpreted autochthonous *Isognomon*
276 banks as indicating a restricted salinity environment, while *P. pustulosa* could tolerate
277 reduced salinity brachy- to mesohaline conditions. Formerly aragonitic gastropod shells are
278 sparite-replaced or, less commonly, neomorphosed; pyrite in a sparite replaced and infilled
279 shell may indicate pyrite growth during or after sparite growth (Fig. 5C).

280

281 In summary, the evidence of frequent exposure and microbial laminites, with a few
282 grainstones, suggests fluctuating energy levels of a very shallow water-body. There is
283 presence of a limited range of more marine-influenced indicators such as forams
284 (agglutinated forms, which range from average-marine to lower salinities) and dasyclads
285 (typical of marine lagoonal settings, from near normal-marine to more restricted conditions;
286 in particular, *B. lusitanica* is found also at very restricted, brackish lagoon facies of the
287 Cabaços Formation in the Vale de Ventos, Serra dos Candeeiros region (Fig. 1B), which

288 suggests it was more tolerant to slight salinity fluctuations than usually acknowledged for
289 dasyclads (Azerêdo et al., 2002a). The bivalves indicate restricted, brachy- to mesohaline
290 salinities. The aforementioned markers are coupled with less common non-marine
291 indicators such as charophyte stems/gyrogonites and changes in the ostracod biota,
292 suggesting fluctuating salinities of a brackish setting, with at times a more marine influence.

293

294 Molluscan remains (Fig. 5) consist mainly of calcitic shells or shell layers, and sparite
295 replacements or less common neomorphic replacements of aragonite by calcite. The latter
296 indicate that the original aragonite was preserved through the TAZ, being later replaced
297 during subsequent burial and congruent dissolution by calcite cement. The preservation of
298 the former aragonitic dasyclad *Barattoloporellopsis* also supports this interpretation but the
299 sparite crystals do not exhibit a pattern of pore-infilling precipitated material, therefore
300 reinforcing the assumption that they are a neomorphic replacement of the original
301 aragonitic coating of the dasyclad (Granier et al. 2017). Minor occurrences of other
302 neomorphically replaced former aragonitic shells also suggest that some aragonite passed
303 through the TAZ. The presence of some pyrite within the sparite-replaced walls and sparite
304 infill of the body chambers in some gastropods (Fig. 5C) suggests sulphate reduction took
305 place but probably after the aragonite had passed through the TAZ.

306

307 2.1.3. Unit 3

308

309 This unit, some 6 m thick (Fig. 6), consists predominantly of matrix-rich limestones,
310 commonly fossiliferous, interbedded with a few laminites, thin marly layers and evaporites
311 at three levels; evidence of exposure is weaker, though rare fenestrae and two pedogenic

312 horizons occur. Porostromate textures, including reworked examples are present, together
313 with coated grains, reworked microbial structures, microbial boundstones and peloidal-
314 intraclastic lenses.

315

316 The ostracod assemblage is of lower diversity and indicates variably brackish to saline
317 conditions, as it lacks the oligohaline *Theriosynoecum*, whereas the euryhaline *S.*
318 *pedrogaensis* is the dominant form and showing differences in size and carapace surface
319 features. The fresh/brackish form *Darwinula* occurs, and locally the euryhaline marine genus
320 *Macrodentina* (*Galliaecytheridea*? in Azerêdo and Cabral, 2004), together with *Klieana*?
321 Dasyclad remains are less common but a low diversity charophyte gyrogonite assemblage is
322 present (Pereira et al., 2003; Azerêdo and Cabral, 2004), whereas charophyte stems are
323 rare. *Botryococcus* while present is absent towards the top of the unit (Barron et al., 1999;
324 Barron and Azerêdo, 2003). The foram *Pseudocyclamina* and a few other agglutinated
325 forms occur and rare echinoid remains are found.

326

327 A particularly prominent, c. 30 cm thick shell-bed exhibits abundant spar-replaced shells
328 of deep-burrowing aragonitic bivalves (*Pholadomya*) in imbricated concentrations and in
329 situ (Fig 6A) with bioclasts forming either planar or cross laminations, the latter mainly
330 towards the top of the bed. Shelly material makes sub-layers within the bed, with sharp
331 erosional or gradational bases. Many of the shells were apparently exhumed and reworked,
332 redeposited and rapidly buried locally, with little fragmentation, but associated in situ shells
333 suggest this was followed by some recolonization.

334

335 Unit 3 shows a marked range of salinity indicators such as forams, rare echinoid debris,
336 coupled with the dasyclads, thus suggesting restricted lagoonal marine conditions. The shell
337 beds are interpreted as the product of storm events.

338

339 Overall, aragonitic molluscs preserved by sparite replacement are uncommon (Fig. 6A, B)
340 although exceptions include one bed rich in very small gastropods close above an evaporite
341 horizon, and the imbricated shell horizon (Fig. 6A). Filament material is less rare, although
342 not common (Fig. 6C).

343

344 2.1.4. Unit 4

345

346 Unit 4 (20 m thick) includes prominent microbial laminites (Fig. 7A), often bituminous
347 and evaporite layers. Oolitic/intraclastic packstones and grainstones are present at the top
348 of the section. There is evidence of some pedogenesis, represented by nodulization fabrics,
349 circumgranular and irregular dissolution/brecciation cracks, and rare very thin carbonate
350 crusts resembling calcicutans.

351

352 This unit has most abundant ostracods, like the previous units, but almost exclusively of
353 the euryhaline form *Sinuocythere pedrogaensis* with larger sized and reticulate carapaces
354 (Colin et al., 2000; Azerêdo and Cabral, 2004). This species even forms coquinas within
355 laminites and is locally closely associated with evaporites. Forams are more common
356 including *Pseudocyclamina parvula* Hottinger, which is known to increase upwards into
357 the overlying marine Montejunto Formation (Ramalho, 1981; Azerêdo et al., 2002a, b). Rare
358 echinoid fragments occur also. Dasyclads are rare and disappear near the base of the unit;

359 charophyte remains are less abundant, more so upwards and show the lowest diversity
360 (Pereira et al., 2003), and the palynomorphs are of lower diversity and marine influenced
361 (Barron and Azerêdo, 2003). Typical marine trace fossils occur locally at the lower part of
362 the unit and include *Rhizocorallium* and *Thalassinoides*. Rare echinoid fragments occur also.
363 Molluscan shell material is predominantly of filaments with occasional steinkerns of small
364 gastropods (Fig.7B) and rare neomorphosed aragonite shells.

365

366 This unit is interpreted as representing shallow, margino-littoral deposition under highly
367 variably hypersaline to marine-influenced conditions, becoming less hypersaline upwards.
368 The lack of sparitic preservation suggests former molluscan aragonite did not survive the
369 TAZ but the filament debris indicates their former presence. This may also explain the
370 decline on preservation of the dasyclads although that could also reflect ecological factors.

371

372 2.2. Taphonomic interpretation

373

374 The Pedrógão succession shows a broad relationship (Fig. 8) whereby former aragonitic
375 shells were initially preserved in fresh to brackish settings from early dissolution in the TAZ
376 but dissolved and filled with calcite cement during later diagenesis (Units 1 and 2). This
377 applies to bimineralic forms as well as those with a wholly aragonitic original shell. Sparite
378 replacement preservation declines through Unit 3, where evaporites become more
379 common. In Unit 4, which represents marine to hypersaline settings the aragonite was
380 preferentially removed syndepositionally leaving only originally calcitic forms, the filaments
381 of calcitic layers of bimineralic forms or steinkerns. However, the pattern is not always
382 consistent. Thin filaments representing only the calcitic layers of bimineralic forms are also

383 common in some beds in Unit 1, suggesting that some aragonite dissolution had occurred,
384 but the filament material is generally seen as very small fragments which could have been
385 transported in from other settings. Overall there appears to have been a reduced level of
386 early, syngedimentary dissolution compared with the overlying units. The former aragonitic
387 dasyclad *Barattoloporellopsis* is also most commonly found in Unit 2, suggesting it was
388 tolerant of lower salinities like some extant dasyclads but its rarity in Units 3 and 4 might
389 largely be an ecological not taphonomic effect.

390

391 Additional factors influence the survival of aragonitic shells past the effects of the TAZ
392 leading either to their continued preservation as aragonite, or to the replacement of
393 aragonite by calcite, as reviewed by Cherns et al. (2008). For example, in high energy
394 deposits including coquinas, fine organic matter is less likely to accumulate, when coupled
395 with the likelihood of more oxygenated sediment, this reduces the amount of sulphate
396 reduction and hence of aragonite dissolution due to sulphide oxidation. A common
397 situation where aragonite can be preserved from early dissolution is in event (storm) beds
398 where rapid burial emplaces the aragonite below the TAZ and many such examples are
399 known (Cherns et al., 2008). This may explain the preservation of the former aragonitic
400 bivalves in Unit 3 which appear imbricated and may have been reworked and rapidly buried
401 (Fig. 6A). Some of these shells are in life position and deeper burrowing forms may be
402 predisposed to have the aragonite initially preserved as they live below the main part of the
403 TAZ (Wright et al., 2003). De Francesco et al. (2013), from assemblages in Holocene lakes in
404 Argentina, ascribed better preservation of molluscs from brackish rather than freshwater
405 lacustrine settings to differences in calcium carbonate saturation. Whether shell thickness
406 played a role in the differential preservation across the salinity gradient is not possible to

407 determine as the aragonitic component in the marine-hypersaline units has been removed.
408 Another consideration is that the availability of Fe in the more argillaceous facies (such as
409 much of Unit 1) might have led to a reduction in dissolution in the TAZ (e.g. Kidwell et al.,
410 2005).

411 The availability of Fe is evidenced by the fact that pyrite is found throughout the four
412 units although is highly variable in abundance. However, sparitic mollusc shells are
413 commonly present in the less argillaceous limestones in Units 1 and 2.

414

415 With the evidence of a significant difference in early aragonite preservation having
416 affected the Cabaços Formation at Pedrógão, and reflecting salinity changes and marine
417 influence, a likely explanation is that sulphate availability was a critical factor. The
418 concentration of sulphate in freshwater can be three orders of magnitude lower than in
419 seawater (Holmer and Storkholm, 2001), limiting the potential production of H₂S as the
420 main agent for triggering aragonite undersaturation and dissolution. This effect is somewhat
421 more complicated as sulphate reducing bacteria, the presumed main source of H₂S in
422 shallow buried sediments occur at similar concentrations in freshwater settings to marine
423 ones (Jørgensen and Bak, 1991; Holmer and Storkholm, 2001). Sulphate usually only occurs
424 to <10 cm into freshwater sediments (e.g. Cook and Schindler, 1983; Holmer and Storkholm,
425 2001), and as a result the zone with the most active sulphate reduction is found in the top
426 0.1 m of sediment (Ingvorsen et al., 1981; Sass et al., 1997), which is in contrast to marine
427 sediments where sulphate reduction can be deeper, although sulphate reduction is often
428 intense despite the low sulphate concentration (Holmer and Storkholm, 2001). Cristini and
429 De Francesco (2017), from a study of Holocene lakes from Argentina, note that the top 10
430 cm of the sediment column is less favourable for molluscan preservation. Although data are

431 limited, they suggest this reduced thickness of the TAZ compared with marine sediment
432 might relate to the role of deeper burrowers in marine settings. The depth of burrowing in
433 lake sediments where bottom waters are not anoxic is much shallower than in marine
434 systems (Hasiotis et al., 2012), generally within the uppermost 0.3 m compared with a
435 maximum range of 1-4 m in marine settings. Thus compared with low energy marine
436 settings the TAZ in lakes should be thinner with skeletal aragonite potentially having a
437 shorter residence time in the TAZ than in a marine setting, making it more likely to survive
438 the zone of aragonite dissolution, and also more readily buried below the TAZ during a
439 storm event. This raises the question as to whether aragonite preservation is more sensitive
440 to deposition rate in non-marine, low sulphate settings.

441

442 3. Discussion

443

444 This study highlights the possible significant preservational differences in molluscs
445 between limestones from freshwater to marine settings, but also shows the range of other
446 factors besides sulphate concentrations that could be involved. There are testable
447 implications arising from this proposal that early diagenesis differs in low sulphate waters.
448 Firstly, former aragonitic molluscs should be more commonly preserved in low energy, low
449 sulphate settings, especially aragonitic shallow-burrowing infaunal bivalves, along with
450 gastropods, which are typically those most readily removed from marine low energy
451 deposits as a result of aragonite dissolution in the TAZ (Cherns and Wright, 2009). Related to
452 this should be the preservation of juvenile forms and micromolluscs (Cherns and Wright,
453 2011). As carbonate released by skeletal aragonite dissolution is the source of the carbonate

454 forming diagenetic nodules and bedding, these might be less common in low sulphate
455 settings if early molluscan aragonite dissolution is much reduced. However, this would also
456 be the case with the alternative proposal (Munnecke and Westphal, 2005) that aragonitic
457 mud is the main source of the diagenetic calcite since this would be less likely in freshwater
458 settings where aragonite producers such as calcareous algae are absent and where
459 precipitation directly from the water column is less likely in low salinity lake waters. During
460 the early Palaeozoic the TAZ was also very thin and as a result the location of cementation
461 below the TAZ was also very near the sediment surface resulting in the widespread
462 occurrence of reworked cemented material as flat pebble conglomerates (Wright and
463 Cherns, 2016). Similar conglomerates do not seem to be a feature of sub-littoral lake
464 deposits and were not noted in the units described in this study.

465

466 As a consequence of the TAZ being thin, perhaps even <10 cm, in low energy lacustrine
467 settings, rapid burial might favour more common aragonite preservation in thin event beds
468 in such low sulphate settings. While it would be difficult to assess the relative frequency of
469 event beds between marine and non-marine successions there are certainly examples of
470 shell beds in non-marine successions. Radley (2009) provides many examples of thin shell
471 beds preserved in Mesozoic brackish-to lacustrine successions, but favours “a fundamental
472 biological control on their genesis; namely small shell size, high reproductive rates and
473 adoption of conservative life modes” (p. 715), and the role of reduced sulphate levels in
474 such settings is not considered. Pan et al. (2012) provide a detailed analysis of lacustrine
475 shell beds from the early Cretaceous Yixian Formation at Sihetun, Beipiao, PRC, focussing on
476 biological and stratigraphic effects not on preservation.

477 Complex patterns of aragonitic molluscan preservation, including the survival of
478 aragonite, are recorded from the late Jurassic-early Cretaceous 'Purbeckian' facies of
479 southern England by El-Shahat and West (1983), representing brackish lagoons. Skawina
480 (2013) provides one of the most detailed analyses of bivalve taphonomy in a lacustrine,
481 charophyte-bearing succession, from the late Triassic of Poland, noting the disarticulated
482 unionoids (originally aragonitic) valves showed no signs of dissolution, interpreting this as
483 evidence the dissolution of the aragonite was a late diagenetic event.

484 The most extensive lacustrine molluscan coquinas, of early Cretaceous age, occur in the
485 South Atlantic. These are documented from the offshore Brazil in the Coqueiros Formation
486 (Lagoa Feia Formation) of Campos Basin (Carvalho et al., 2000; Thompson et al., 2015), the
487 Itapema Formation of Santos Basin (Carlotto et al., 2017), and the Sergipe-Alagoas Basin in the
488 Morro de Chaves Formation (Kinoshita, 2010; Tavares et al., 2015), and offshore West Africa
489 in the 'Toca' (e.g. the Congo and Cabinda basins; Harris, 2000). In the Campos Basin the
490 Barremian coquinas are associated with fluvial-alluvial deposits and lacustrine carbonate
491 bodies, deposited in a complex lake system controlled by local environmental conditions
492 and tectonics. Its thickness varies from 2,400 m in palaeostructural lows to 0 m in
493 palaeostructural highs. The main hydrocarbon reservoir type consists of coquina lenses
494 characterized by units over 10 m thick made up of amalgamation of relatively thin layers
495 (0.20–1 m thick) of bivalve-rich calcirudites, classified as grainstones, that locally show cross
496 stratification, associated with thin layers of matrix-poor and matrix-dominated carbonates
497 and no siliciclastic rock intercalations. Successions comprised of this facies sub-association
498 form layers 20–550 m thick. The sedimentary features suggest high energy and shallow
499 water depositional conditions. Although many of the coquina deposits represent high
500 energy shoreline facies others are associated with low energy matrices and the contrast

501 with their marine equivalents where infaunal, former aragonitic bivalves are only rarely
502 preserved might be explained by the lower sulphate content of the lake waters. However,
503 non-marine systems are highly varied and dynamic in terms of their chemistries and many other
504 diagenetic pathways are possible for molluscan skeletal preservation (Fursich and Pan, 2016).

505

506 **4. Conclusions**

507

508 A fundamental difference should exist between the early diagenesis of aragonite in
509 marine and freshwater environments because of the degrees of sulphide activity and its
510 effect on aragonite preservation. Growing evidence shows that aragonite is lost, effectively
511 syndepositionally, from low energy marine sediments because of undersaturation triggered
512 by oxidation of H₂S generated by sulphate-reducing bacteria. Such effects should be reduced
513 in sulphate-poor settings such as freshwater lakes. To test this hypothesis a range of fresh-
514 brackish-hypersaline and marine limestones was sampled from the Upper Jurassic (mid-late
515 Oxfordian) Cabaços Formation of central western Portugal. In limestones deposited in
516 freshwater to brackish settings many molluscs, mainly gastropods, are preserved as sparite
517 shell replacements indicating that the original aragonite was preserved through the TAZ but
518 were later replaced during subsequent burial by calcite cement. In limestones representing
519 deposition in more marine to higher salinity settings molluscan remains mainly consist of the
520 calcitic layers of bimineralic bivalves, or of calcitic shells, or gastropod steinkerns. While
521 there seems to be dominant diagenetic/taphonomic preservation patterns related to the
522 two main facies associations (fresh/fresh-brackish water and marine-hypersaline), a
523 consistent pattern is not always present, reflecting the high-frequency salinity changes
524 recorded in the Cabaços Formation. This potential preservational bias in low sulphate waters

525 towards aragonitic fossils requires further study and might explain the anomalous thickness
526 and abundance of some lacustrine molluscan limestones such as those making significant
527 hydrocarbon reservoirs in the South Atlantic.

528

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704

705 **Figure captions**

706 Fig. 1. A, inset map of the Iberian Peninsula indicating study area in western Portugal; B, simplified
707 geological map of the onshore part of the Lusitanian Basin, western central Portugal showing
708 outcrop area of Jurassic limestones, with location of Pedrogão and other key localities in bold.

709 Fig. 2. Simplified stratigraphical framework (not to scale) for the Middle–early Upper Jurassic of
710 the Lusitanian Basin (Cabaços Formation shaded) showing the interpretation of dominant
711 depositional environments/facies types.

712 Fig. 3. Log of the section at Pedrógão showing the stratigraphic distribution of sedimentary features,
713 key biota and types of molluscan replacement. Note that while ostracods are ubiquitous in almost all
714 levels, the associations are salinity-sensitive so composition and proportions of taxa differ (see text),
715 which is summarized by marking distribution as two broad groups: i) freshwater/brackish and ii)
716 euryhaline forms. When both groups occur, the dominant one is indicated by a square around the
717 bullet. Four facies/palaeoecological subdivisions recognised by Azerêdo and Cabral (2004) from
718 a range of palaeontological (chiefly the ostracod associations) and sedimentological data are
719 here labelled Units 1-4, updated and taken as independent background to test our
720 interpretations.

721 Fig. 4. Lithofacies Unit 1 at Pedrógão. A, calcite encrusted charophyte stems in coquina near the base
722 of the Cabaços Formation; B, gastropod shells, some partially replaced by sparite and some with
723 thin, well preserved calcitic outer layer (residue retrieved from >0.5mm fraction of a marl sample
724 studied for microfossils); C, fragments of well-preserved outer calcitic layer of a bimineralic
725 gastropod shell, and a well preserved calcitic ostracod carapace.

726 Fig. 5. Lithofacies Unit 2 at Pedrógão. A-B, *Isognomon* shell bed, shells mainly articulated, with well
727 preserved calcitic outer shell layer; C, gastropod with sparite walls and infill of the body chamber
728 including some pyrite, which suggests pyrite formation during or after sparite growth.

729 Fig. 6. Lithofacies Unit 3 at Pedrógão. A, imbricated pholadomyacean shells with sparite-replaced
730 walls; B, sparite-replaced former aragonitic gastropod shell and *B. lusitanica* (dasyclad); C, 'filaments'
731 representing calcitic outer layer(s) of bivalve shell, inner aragonitic layers have been dissolved.

732 Fig. 7. Lithofacies Unit 4 at Pedrógão. A, microbial laminites; B, steinkern of gastropod, suggesting
733 early dissolution of formerly aragonitic walls, associated with ostracods and a charophyte
734 gyrogonite.

735 Fig. 8. The effect of salinity on the potential for aragonite shell preservation. In fresh to brackish (low
736 sulphate) waters sulphide activity is limited in the TAZ (Taphonomically Active Zone), increasing the
737 potential for initial aragonite preservation, later to be replaced by sparite in the FBZ (final burial
738 zone). In more sulphate-rich waters aragonite is normally preferentially removed and moulds are
739 typically destroyed by bioturbation; filaments representing the outer prismatic calcite shell layer of
740 bimineralic bivalves and steinkerns may be preserved.

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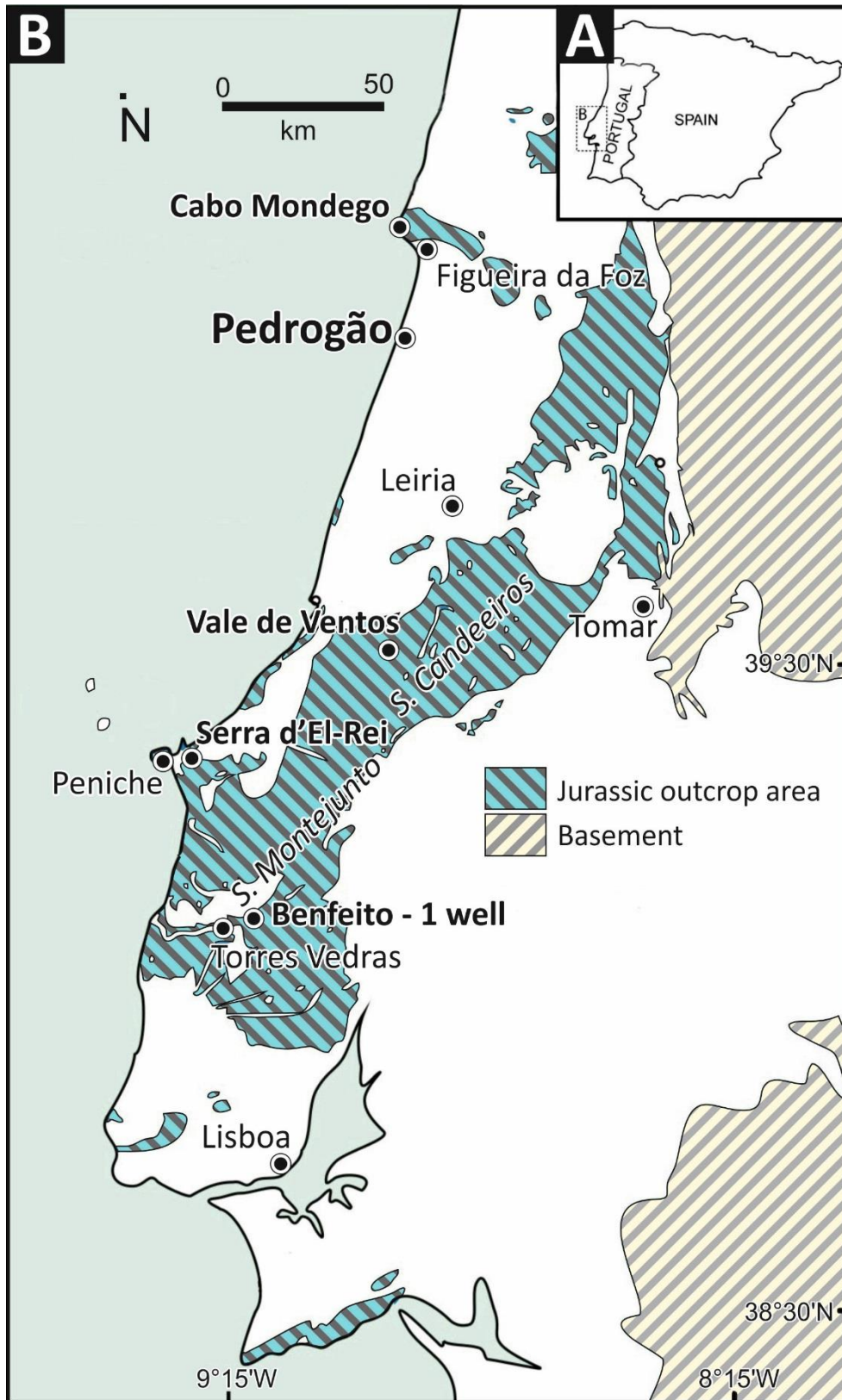
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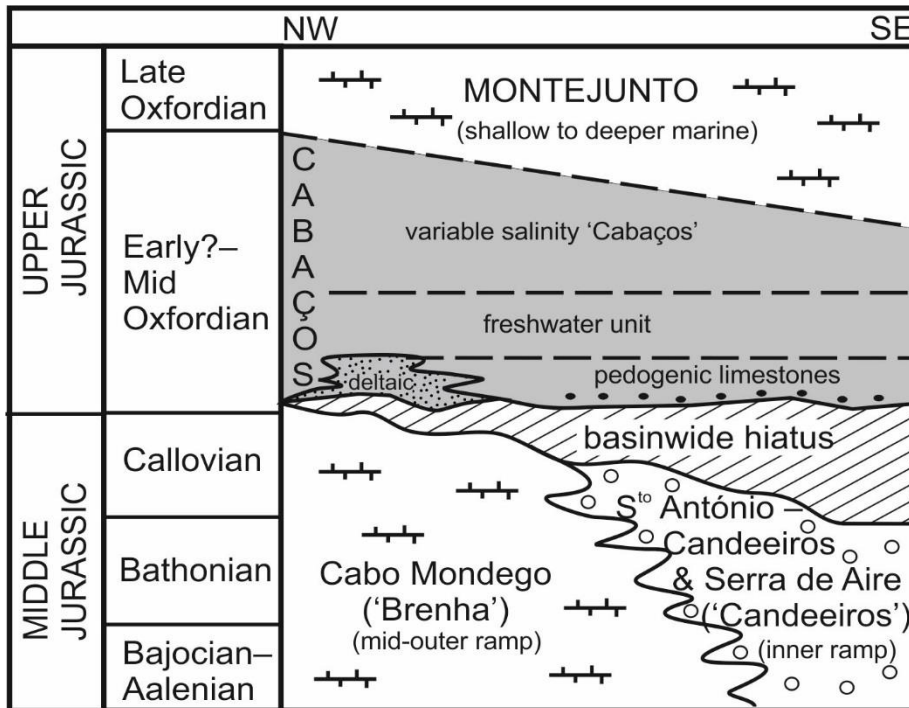
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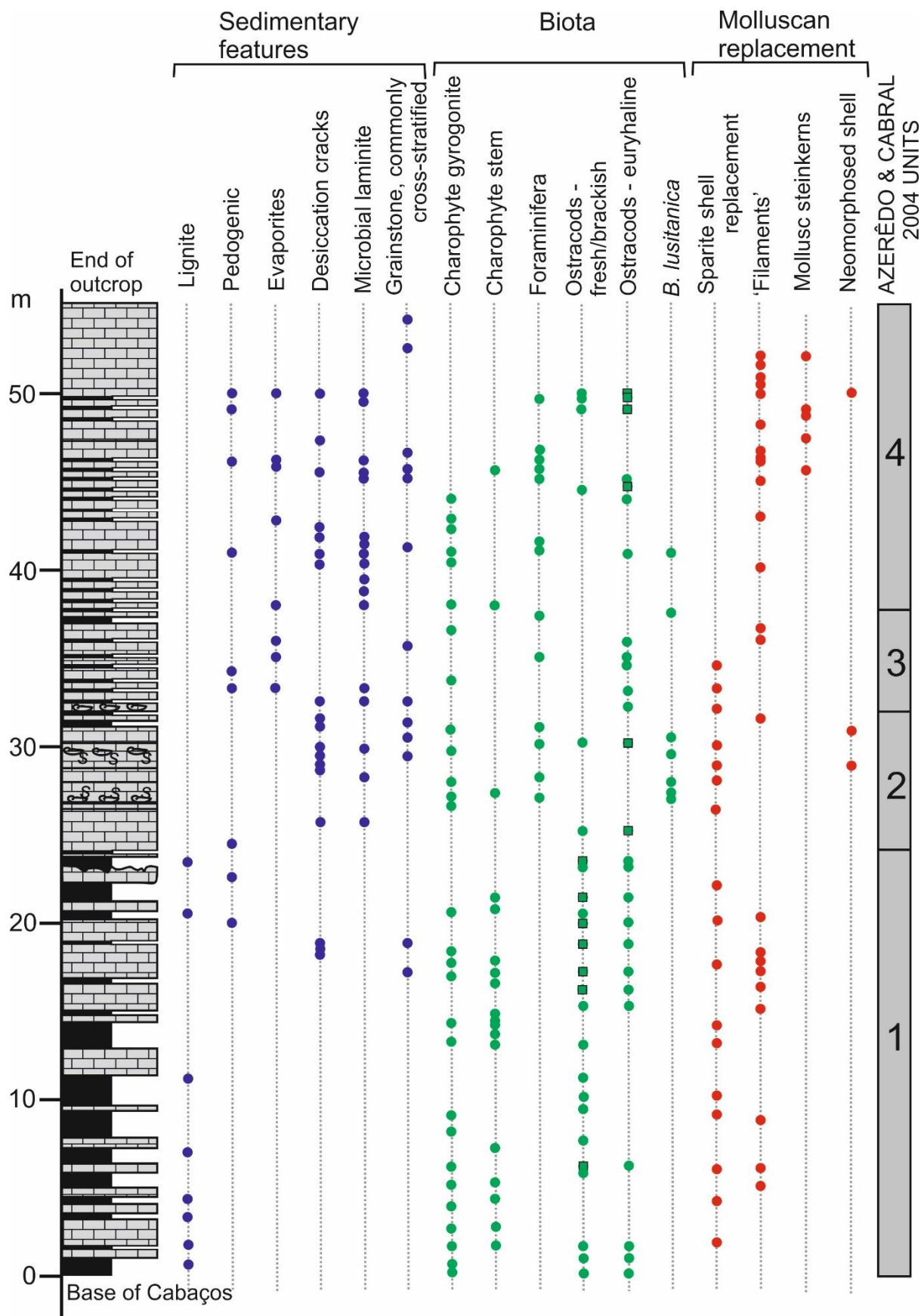
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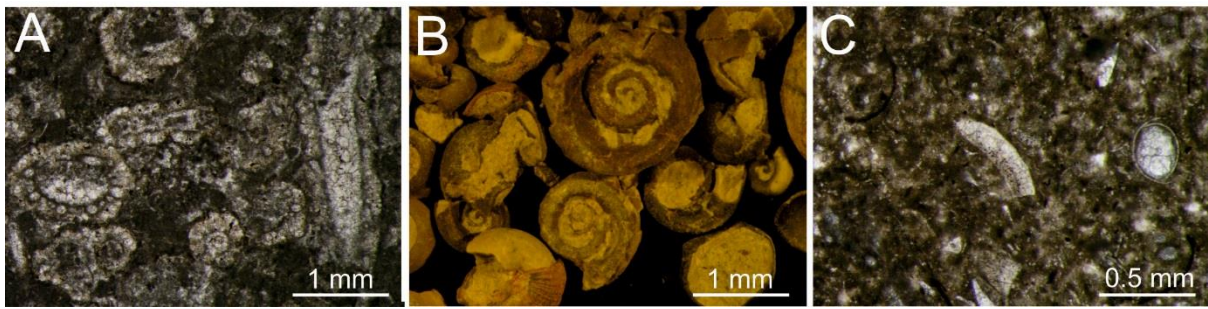
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760 Figure 4



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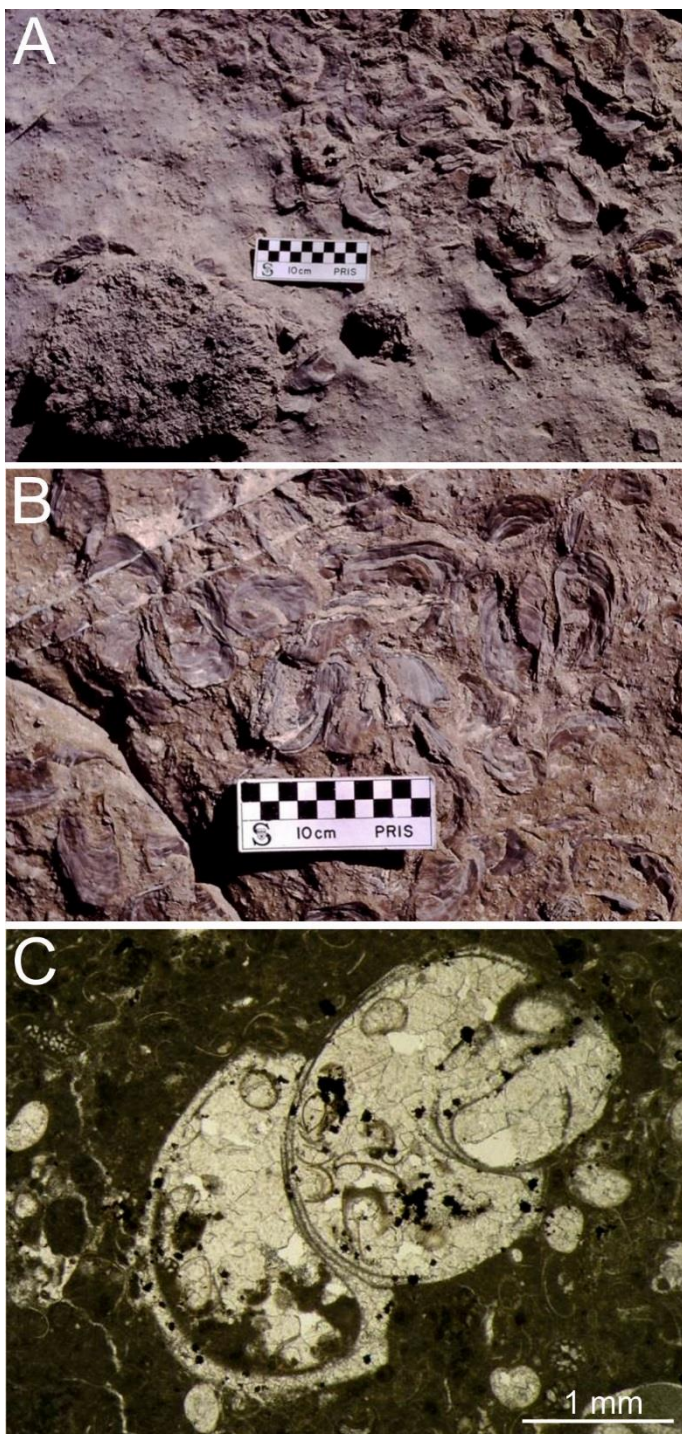
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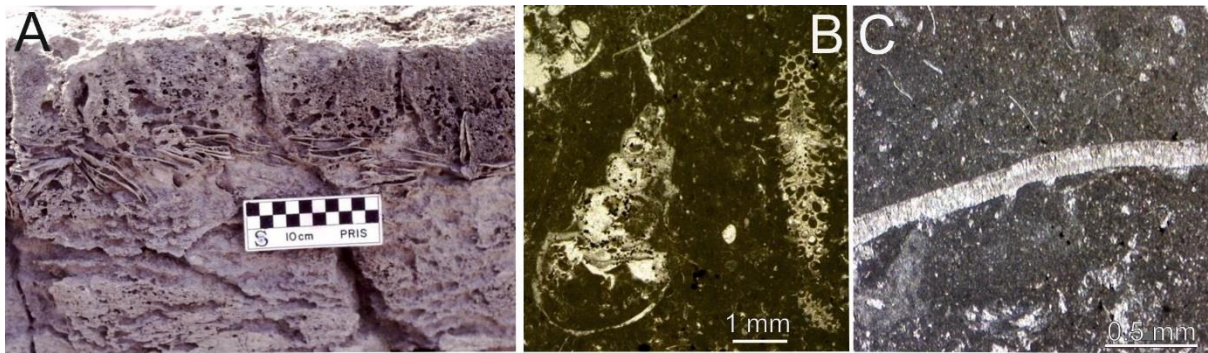
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778 Figure 5



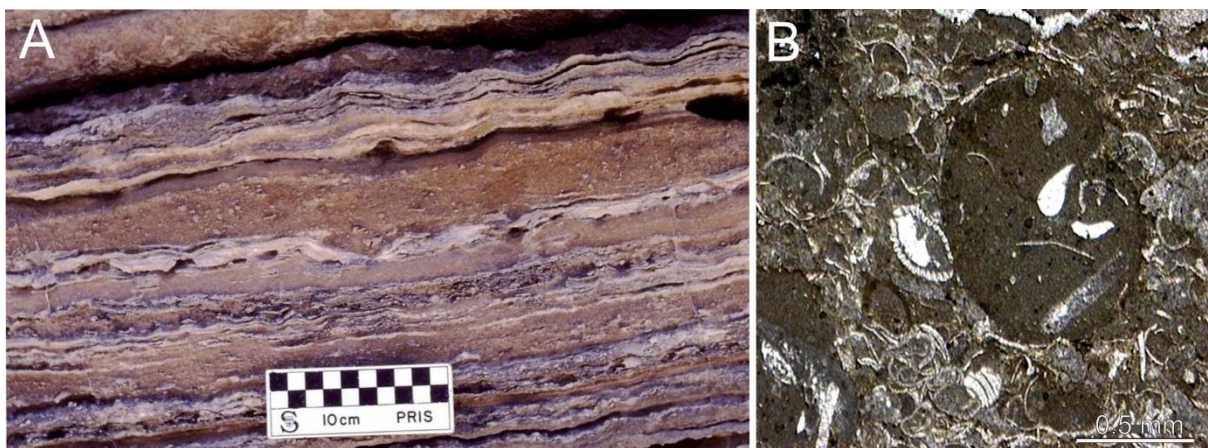
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780 Figure 6



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782 Figure 7



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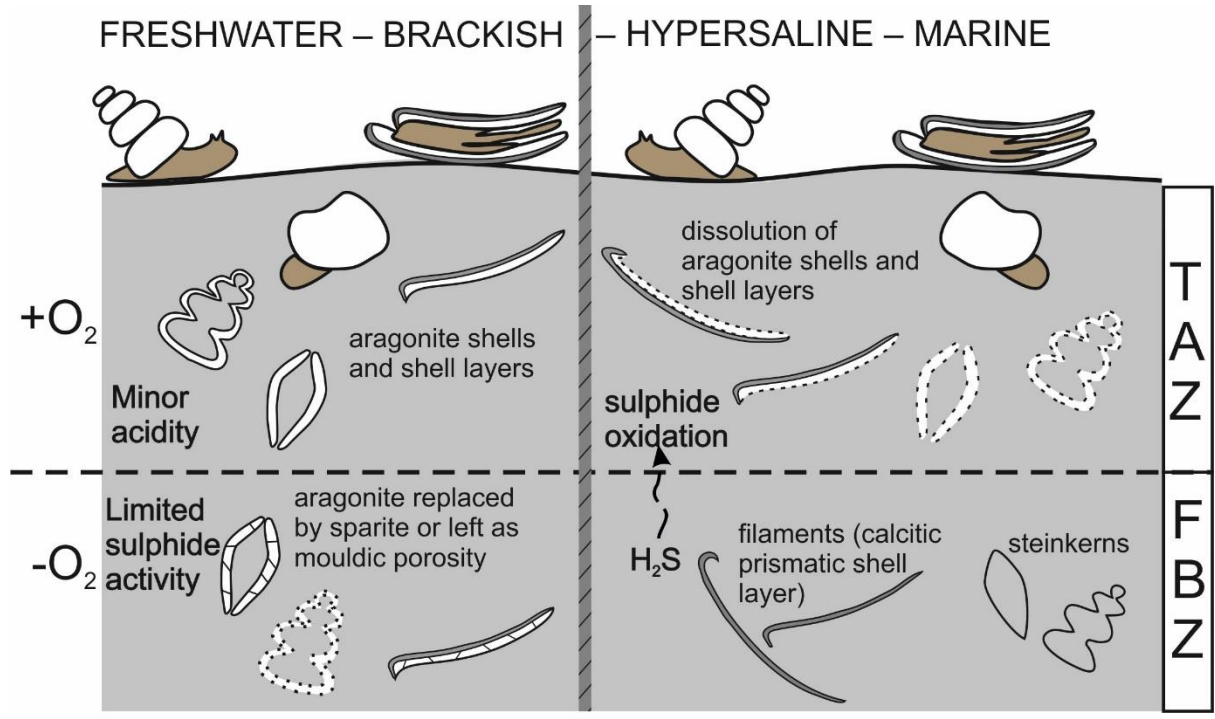
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792 Figure 8



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