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1 **Carbonaceous microstructures from sedimentary laminated chert within the**  
2 **3.46 Ga Apex Basalt, Chinaman Creek locality, Pilbara, Western Australia**

3

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28

29 **Abstract**

30 Hydrothermal black chert veins intruding the 3.46 Ga Apex Basalt contain some of  
31 Earth's oldest microfossil-like objects, whose biogenicity has been questioned. Whilst  
32 these black chert veins have been studied in great detail, relatively little is known  
33 about the stratiform, seafloor, sedimentary cherts that are conformably interbedded  
34 with volcanic rocks of the Apex Basalt.

35

36 Herein, we document and assess the biogenicity of carbonaceous microstructures  
37 present in the lowermost of the stratiform chert units (informally known as the 'Apex  
38 chert'), at the Chinaman Creek locality in the Marble Bar greenstone belt, Pilbara  
39 Craton, Western Australia. Carbonaceous material mostly occurs within clotted grey-  
40 black cherts and microgranular 'grainstone-like' cherts within the stratiform unit, the  
41 latter being the major focus of this study. In the clotted cherts, carbon occurs as  
42 lobate, fluffy grains, rare compressed flakes, and as a grain boundary phase around  
43 spherulitic silica. There is no morphological evidence to support the biogenicity of  
44 these microstructures. In contrast, the microgranular chert contains fluffy and flaky  
45 carbonaceous grains, plus laminated grains comprising multiple non-isopachous  
46 wrinkled carbonaceous laminae, with noted thickening towards some ridge crests, as  
47 determined by confocal laser scanning microscopy. Roll-up structures provide  
48 evidence of an initial plasticity, interpreted to have formed via the tearing-up and  
49 current-induced plastic deformation of microbial mat fragments. Geochemical  
50 mapping, using laser Raman micro-spectroscopy and NanoSIMS, respectively

51 demonstrates the antiquity of the carbon, and reveals a close correlation between  
52 carbon, nitrogen and sometimes sulfur, concentrated within dark brown to black  
53 laminae. Adjacent to microgranular zones are zones of more persistent carbonaceous,  
54 undulose, filament-like laminae that entrain relict sediment grains. These  
55 microstructures are directly comparable to a sub-type of microbially induced  
56 sedimentary structure (MISS), widely reported from younger siliciclastic sediments  
57 colonized by microbial biofilms.

58

59 The morphology and chemical composition of both the non-isopachous laminated  
60 grains and the filament-like laminae are consistent with a biological interpretation,  
61 suggesting microscopic MISS were present in the microgranular stratiform 'Apex  
62 chert'. However, the fact that neither macroscopic MISS nor *bona fide* microfossils  
63 have yet been reported from this unit, coupled with the proximity of these structures  
64 to active hydrothermal vents, potentially discharging hot carbon-rich fluids, urges  
65 caution in such an interpretation. The Chinaman Creek 'Apex chert' investigated here  
66 is one of at least five sedimentary, laminated cherts within the Apex Basalt. These  
67 horizons are promising targets in the search for biological activity within a  
68 dominantly volcanic Archean environment.

69

70 **Keywords:** Apex chert; Pilbara; Carbon; MISS; Archean life

71

## 72 **1. Introduction**

73 The 3.46 Ga Apex Basalt in the Marble Bar greenstone belt of the Pilbara Craton,  
74 Western Australia, has long been a focus for the investigation of early Earth  
75 environments and potential microbial life. It is particularly notable for the presence of

76 carbonaceous filamentous microstructures in hydrothermal black chert veins at the  
77 Chinaman Creek locality, interpreted as representing eleven species of fossilized  
78 prokaryotes (Schopf and Packer, 1987; Schopf, 1992, 1993, 1999). However, the  
79 biogenicity of these ‘microfossils’ has been highly debated (e.g., Brasier et al., 2002,  
80 2005, 2011; Schopf et al., 2002, 2007, 2010; Schopf and Kudryavtsev, 2009, 2012,  
81 2013; Marshall et al., 2011; Ollcott Marshall and Marshall, 2013). Recent  
82 morphological and geochemical analyses at a high spatial resolution have shown the  
83 filaments to be mineral artifacts comprising chains of phyllosilicate crystals that later  
84 adsorbed carbon during fluid flow within an active hydrothermal system (Brasier et  
85 al., 2015; Wacey et al., 2015), or carbon-filled cracks (Bower et al., 2016). Biogenic  
86 stromatolitic clasts have also been postulated in the Apex hydrothermal chert veins  
87 (Schopf, 1993), though these were subsequently reappraised as isopachous, abiogenic,  
88 stromatoloidal internal cements, occurring in a later stage chert fabric (fabric B2 of  
89 Brasier et al., 2005).

90  
91 Despite the controversy over the presence of life in the Apex hydrothermal chert  
92 veins, the Apex Basalt remains a promising rock unit to investigate early life on Earth.  
93 It is well-preserved, having undergone metamorphism to no greater extent than  
94 prehnite-pumpellyite to lower greenschist facies (Hickman and Lipple, 1978; Van  
95 Kranendonk et al., 2007). It also contains at least five stratiform chert units (Kato and  
96 Nakamura, 2003; Van Kranendonk, 2006); these are concordant, bedding parallel  
97 units that are often internally laminated and preserve sedimentary structures (Kato and  
98 Nakamura, 2003; Brasier et al., 2011). Hence, these may provide evidence of  
99 sedimentary environments (and their associated biotas) reflecting quiescent periods in  
100 an otherwise volcanic ‘Apex time’. In some recent studies it has proven effective to

101 look in volcanically influenced terranes for signs of early life (e.g., Walsh, 2004;  
102 Westall et al., 2006, 2015), since these provide many of the minor elements essential  
103 for life (e.g. Cu, Co, Ni and Fe; Barras, 2012). Volcanic lithologies have shown the  
104 capability to foster modern extremophilic life; shortly following their eruption,  
105 bacterial communities are able to benefit from the diverse, often metallic, elements  
106 present (Cockell et al., 2009; Kelly et al., 2011, 2014).

107

108 Here, we investigate the lowermost of the stratiform chert horizons of the Apex Basalt  
109 at the Chinaman Creek locality (Unit 4 of Brasier et al., 2005, 2011; informally  
110 referred to as the ‘Apex chert’), outlining the modes of occurrence of carbonaceous  
111 material, and assessing the likelihood of a biogenic component. In so doing, we  
112 consider that very ancient/alien putative biogenic structures and geochemical signals  
113 should not be accepted as being of biological origin without geologically-plausible  
114 non-biological origins first being tested and falsified (cf. Brasier et al., 2004a).

115

## 116 **2. Methods**

### 117 *2.1. Field mapping and petrographic analysis*

118 Field mapping in the Marble Bar greenstone belt was undertaken by us as part of a  
119 wider programme with the Geological Survey of Western Australia, supplemented by  
120 a detailed programme of mapping and sampling of the Apex Basalt across an area of  
121 around 12 km<sup>2</sup>. Multiple samples were collected from the Chinaman Creek locality  
122 between 1999 and 2006, located by means of satellite images and Global Positioning  
123 Systems (GPS). For the stratiform ‘Apex chert’, samples were collected across the full  
124 1.5 km of available outcrop, encompassing each of the north, central and south blocks  
125 described in Brasier et al. (2005, 2011). Optical petrography and fabric mapping was

126 performed in the Department of Earth Sciences imaging laboratory, Oxford  
127 University, using standard 30  $\mu\text{m}$  and 100  $\mu\text{m}$  petrographic thin sections. Thin  
128 sections were examined under bright-field, polarized transmitted, and incident  
129 (reflected) light using Nikon Optiophot-2 (biological) and Optiophot-pol (polarizing)  
130 microscopes. Images were obtained using a single-chip CCD camera, providing live  
131 images in full RGB colour, and processed using AcQuis and Auto-Montage image  
132 capturing software.

133

## 134 *2.2. Confocal Laser Scanning Microscopy (CLSM)*

135 Confocal images were acquired for numerous features observed in thin-sections using  
136 a Nikon A1-Si laser-scanning confocal microscope using either a 20x or 40x oil-  
137 immersion objective (numerical apertures of 0.7 or 1.3, respectively). Images were  
138 recorded with pixel dimensions between 0.31 and 0.61  $\mu\text{m}$ . Autofluorescence of the  
139 specimens was excited with the following laser lines: 405-nm line of 100 mW cube  
140 laser (Coherent Inc., USA, <http://www.coherent.com>), 488-nm line of 50 mW  
141 sapphire laser (Coherent Inc., USA), 561-nm line of 50 mW sapphire laser (Coherent  
142 Inc., USA) and 640-nm line of 40 mW cube laser (Coherent Inc., USA).

143 Autofluorescence signal was collected with 4 PMT detectors with the following  
144 wavelength emission windows: 425–475 nm for the 405 nm laser, 500–550 nm for the  
145 488 nm laser, 570–620 nm for the 561 nm laser, and 675–725 nm for the 640 nm  
146 laser. The specimens were visualised using a 29.9  $\mu\text{m}$  (1.2 airy units) confocal  
147 pinhole and a number of z-stacks (typically between 10 and 50), of optical thickness  
148 between 0.2–2.0  $\mu\text{m}$  each, were acquired. The fluorescence signal from each z-stack  
149 was then projected onto a maximum projection image and used to generate a 3D  
150 model of the specimen using Nikon NIS-Elements software ([www.nis-elements.com](http://www.nis-elements.com))

151 for Figure 13c. The stacks were further explored and visualised using volume  
152 rendering; they were loaded using the open source software Fiji (Schindelin et al.,  
153 2012), and the channel with maximum contrast converted to a grayscale TIFF. The  
154 resulting TIFF stacks were subsequently loaded in the open source software Drishti  
155 (Limaye, 2012), and volume rendered by modifying the 2D histogram transfer  
156 function. These renders were used as the basis for Figure 13b, d and for  
157 supplementary movie 1.

158

### 159 *2.3. Nano-scale Secondary Ion Mass Spectrometry (NanoSIMS)*

160 NanoSIMS was performed in the Department of Materials, University of Oxford,  
161 using a CAMECA NanoSIMS 50. Regions of interest (ROI) were identified under the  
162 optical microscope in polished 30  $\mu\text{m}$  thin sections, and then micro-mapped using  
163 bright-field and reflected light. The reflected light images were subsequently used to  
164 locate the surface expressions of laminated features within the NanoSIMS. Discs of c.  
165 10 mm diameter containing the ROI were extracted from the thin sections, mounted  
166 on NanoSIMS stubs, and coated with a thin (5-10 nm) layer of platinum to provide  
167 conductivity at high voltage. Details of qualitative elemental mapping using  
168 NanoSIMS in multi-collector mode are given in Wacey et al. (2008) and Kilburn and  
169 Wacey (2011). Briefly, a focused primary  $\text{Cs}^+$  ion beam, with a beam current of 2–4  
170 pA, was rastered over the sample surface, and the sputtered ions were extracted to a  
171 double focusing mass spectrometer. Images with sub-100 nm spatial resolution  
172 mapping relative ion intensity were acquired over fields of view ranging from 10  $\mu\text{m}$   
173 to 25  $\mu\text{m}$ . Prior to each analysis, the sample area was pre-sputtered to remove surface  
174 contamination, implant  $\text{Cs}^+$  ions into the sample matrix and attain an approximate  
175 steady state of secondary ion emission (cf. Gnaser, 2003). Ion maps of carbon ( $^{12}\text{C}^-$ ),



176 nitrogen ( $^{12}\text{C}^{14}\text{N}^-$ ), silicon ( $^{28}\text{Si}^-$ ), sulfur ( $^{32}\text{S}^-$ ) and phosphate ( $^{31}\text{P}^{16}\text{O}_2^-$ ) were then  
177 produced simultaneously from the same sputtered volumes of sample. Only relative  
178 concentrations of elements can be obtained using this NanoSIMS methodology.  
179 Without multiple standards, no inferences can be made from these data concerning  
180 either the absolute concentration of elements, or the percentage concentration of one  
181 element compared to another.

182

#### 183 *2.4. Laser Raman Microspectroscopy*

184 Raman was performed in the Centre for Microscopy, Characterisation and Analysis  
185 (CMCA), The University of Western Australia, using a *WITec alpha 300RA+*  
186 instrument with a *Toptica Photonics Xtra II* 785 nm laser source. Laser excitation  
187 intensity at the sample surface was in the 1-5 mW range, well below the intensity that  
188 may damage carbonaceous material (e.g., Everall et al., 1991) and comparable to  
189 previous studies of the Apex chert (e.g., Olcott Marshall et al., 2012; Sforza et al.,  
190 2014). The laser was focused through either a 20x/0.4 or 100x/0.9 objective, with the  
191 latter giving a spot size of smaller than 1  $\mu\text{m}$ . Spectral acquisitions were obtained  
192 with 600 l/mm grating and a peltier-cooled (-60 °C) 1024 x 128 pixel CCD detector.  
193 Laser centering and spectral calibration were performed daily on a silicon chip with  
194 characteristic Si Raman band of 520.4  $\text{cm}^{-1}$ . Count rates were optimised prior to point  
195 spectra acquisition or hyperspectral mapping using the dominant quartz Raman band  
196 of 465  $\text{cm}^{-1}$ . Spectra were collected in the 100-1800  $\text{rel. cm}^{-1}$  region in order that both  
197 1<sup>st</sup> order mineral vibration modes and 1<sup>st</sup> order carbonaceous vibration modes could  
198 be examined simultaneously. Raman maps were acquired with the spectral centre of  
199 the detector adjusted to 944  $\text{cm}^{-1}$ , with a motorised stage allowing XYZ displacement  
200 with precision of better than 1  $\mu\text{m}$ . Spectral decomposition and subsequent image

201 processing were performed using WITec Project FOUR software, with baseline  
202 subtraction using a 3<sup>rd</sup> or 4<sup>th</sup> order polynomial. Carbon maps were created by  
203 integrating over the ~1600 cm<sup>-1</sup> 'G' Raman band and quartz maps created by  
204 integrating over the 465 cm<sup>-1</sup> quartz Raman band. The ~1350 cm<sup>-1</sup> carbon 'D' Raman  
205 band was not used to construct maps because this may suffer from interference from  
206 the ~1320 cm<sup>-1</sup> hematite Raman band (cf. Marshall and Olcott Marshall, 2013). Point  
207 spectra were acquired using the 100x/0.9 objective, an integration time of 0.5 s and 10  
208 accumulations. All analyses were conducted on material embedded below the surface  
209 of the thin section to avoid artefacts in the Raman spectra resulting from polishing  
210 and/or surface contamination.

211

## 212 2.5. Energy dispersive elemental mapping (EDS)

213 Elemental analysis and mapping over several millimeters of Chinaman Creek thin  
214 sections was performed on a *FEI Verios 460* SEM equipped with an *Oxford*  
215 *Instruments X-Max 80* energy dispersive X-ray spectroscopy (EDS) system and  
216 *Oxford Instruments AZtec 3.0* nano-analysis software, located in CMCA.

217

## 218 3. Context

### 219 3.1. Regional setting

220 The c. 3.46 Ga Apex Basalt is found in the East Pilbara terrane of the Pilbara Craton,  
221 Western Australia (Fig. 1). This c. 3.53–3.23 Ga terrane contains some of Earth's  
222 oldest and best-preserved rocks, and comprises a series of domed granitoid  
223 complexes, intruded into and overlain by volcano-sedimentary rocks of the Pilbara  
224 Supergroup (Van Kranendonk et al., 2007; Hickman, 2008, 2012). The Pilbara  
225 Supergroup is divided into three unconformity-bound lithostratigraphic groups

226 (Warrawoona, Kelly, and Sulfur Springs). These crop out across c. 20 greenstone  
227 belts in the East Pilbara, each belt dipping away from the granitoids (Van Kranendonk  
228 et al., 2001; Hickman, 2012). The lowermost of these groups, containing the Apex  
229 Basalt, is the Warrawoona Group, a 10-15 km thick volcano-sedimentary succession  
230 deposited between c. 3.53 and 3.43 Ga, dominated by extrusive volcanic rocks with  
231 minor interstratified chert, barite, carbonate and volcanoclastic units (Hickman, 1983;  
232 Van Kranendonk et al., 2007). The Apex Basalt is best exposed in the Marble Bar  
233 greenstone belt where it is c. 3 km thick; here it overlies the Marble Bar Chert  
234 member of the c. 3.47 Ga Duffer Formation and is in turn overlain by felsic volcanics  
235 of the c. 3.45 Ga Panorama Formation (Van Kranendonk, 2006).

236

### 237 *3.2. Chinaman Creek Geology*

238 In the vicinity of Chinaman Creek, approximately 5 km west of Marble Bar, thick  
239 extrusive accumulations of pillow basalt and komatiite are punctuated by a  
240 weathering-resistant ridge of stratiform chert and associated volcanoclastic rocks,  
241 informally referred to as the 'Apex chert' (Fig. 2). The stratiform chert (unit 4 of  
242 Brasier et al., 2005, 2011), the lowermost and thickest of several stratiform cherts in  
243 the Apex Basalt (Kato and Nakamura, 2003), is a 10-15 m thick unit of banded iron-  
244 rich and iron-poor chert (cf. banded iron formation) of variable texture, composition  
245 and colour. Brasier et al. (2005, 2011) recognized that the stratiform chert ridge was  
246 separated into three structural blocks (naming them the North, Central and South  
247 blocks) by listric normal growth faults. They also showed that, stratigraphically below  
248 the ridge, a series of hydrothermal black chert veins cut up through the lower portions  
249 of the Apex basalt for up to 1600 m (Figs. 2-3). The veins are particularly thick (up to  
250 c. 5 m in diameter) along the growth faults and one such vein (N1 of Brasier et al.,

251 2005; Fig. 3) contains the filamentous microfossil-like artifacts described by Schopf  
252 (1993), at a depth of c. 100 m below the palaeosurface. The black chert veins inter-  
253 finger with, but do not pass through, the upper stratigraphic limit of the stratiform  
254 chert (Brasier et al., 2005; Van Kranendonk, 2006) indicating that they are  
255 syndepositional with, or penecontemporaneous to, this unit (Fig. 3). Large clasts of  
256 both hydrothermal black chert and stratiform chert are found in the overlying  
257 pyroclastic breccia bed (unit 5 of Brasier et al., 2005, 2011) indicating that both were  
258 lithified prior to the commencement of the next volcanic cycle. In the vicinity of these  
259 veins, the stratiform chert can be highly brecciated with dilatational black chert  
260 artificially thickening the stratiform succession and creating angular blocks of bedded  
261 chert that appear to ‘float’ in black chert (Brasier et al., 2005), confirming that at least  
262 some of the stratiform chert must predate the black chert. Stratiform material may also  
263 be found entrained at depth (up to c. 150 m) within the black chert veins, likely  
264 caused by either vigorous downward convection during hydrothermal fluid flow or  
265 phreatomagmatic explosions (Van Kranendonk, 2006). In contrast to most previous  
266 studies that have sought to address the origin of carbonaceous structures in and  
267 around the hydrothermal veins (Wacey et al., 2015; Bower et al., 2016 and references  
268 therein), here we focus on stratiform chert that shows evidence for sedimentary  
269 structures, and mostly crops out some distance away from the major hydrothermal  
270 veins (Fig 3).

271

## 272 **4. Results**

### 273 *4.1. Petrographic division of stratiform chert types*

274 The stratiform chert is present in each of the three structural blocks at Chinaman  
275 Creek, but is most continuous in the north and south blocks. The central block has a

276 comparative paucity of both stratiform and hydrothermal chert. We here divide the  
277 stratiform chert at Chinaman Creek into five distinct types on the basis of  
278 petrographic observations: silicified volcanoclastics (mostly layered ash and  
279 agglomerate; Fig. 4a-b), clotted carbonaceous chert (Fig. 4c-d; cf. Lowe and Knauth  
280 (1977), banded microgranular chert (Fig. 4e-f; Supp. Figs. 1-2), metalliferous, mostly  
281 iron-rich chert (Fig. 4g-h) and banded black, grey and white chert (Fig. 4i-j).

282

283 This study focuses on the banded microgranular cherts, which not only preserve  
284 significant quantities of carbonaceous material, but also show clear sedimentary  
285 textures, such as grain orientation and sorting (Fig. 4e; Supp. Figs. 1-2). This is  
286 particularly apparent for the largest grains in the microgranular chert (Supp. Fig. 2).  
287 Other chert types are currently under detailed investigation but are beyond the scope  
288 of this study. The microgranular cherts provide the widest range of carbonaceous  
289 textures, though some of these are shared with other chert types: for example,  
290 laminated textures on the mesoscale define the fabrics of parts of the microgranular  
291 chert, plus parts of the black, grey and white and iron-rich banded cherts. However,  
292 microscopic laminations within individual grains are solely found within  
293 microgranular cherts. Well-developed spherulitic silica textures characterise the  
294 clotted carbonaceous cherts, but are also minor components of the microgranular and  
295 other cherts.

296

#### 297 *4.2. Microgranular chert fabrics*

298 Fabrics within the microgranular cherts are variable, even on the scale of a single  
299 standard thin section and include (i) microgranular zones, (ii) laminated textures and  
300 (iii) spherulitic textures, which we address in order below. Additionally,  $\mu\text{m}$ - $\text{mm}$  scale

301 post-depositional micro-quartz and macro-quartz veins represents multiple later  
302 episodes of veining. Brecciation of cherts is common around the large hydrothermal  
303 intrusive black chert veins, however, data herein come from non-brecciated stratiform  
304 chert away from macro-scale veins.

305

#### 306 4.2.1. Microgranular zones and grain types

307 Microgranular zones comprise grains of various shapes and sizes that show some  
308 degree of sorting and a preferred orientation (Fig. 4e-f; Supp. Figs. 1-2). These zones  
309 show colour banding in roughly equal proportions of light and dark material (cf.  
310 ‘laminated silty argillites’ in Cressman, 1989; Scheiber, 1990; Scheiber et al., 2012);  
311 optically lighter and darker bands alternate on the sub-millimetre- to millimetre-scale,  
312 and have markedly differing characters (Fig. 4e-f; Supp. Fig. 1). Microgranular cherts  
313 are generally well-sorted and usually grain-supported, though lobate grain-rich layers  
314 are locally matrix-supported, principally through their high interparticle porosity.  
315 Subtle imbrication of grains is present, especially in elongate flakes. This lithology  
316 superficially resembles a fine-grained, shallow-water grainstone or pelsparite, though  
317 is more compositionally akin to a silicified shale (Schieber et al., 1990).

318

319 The most common components of the microgranular chert are sub-rounded,  
320 carbonaceous lobate grains or ‘fluffs’ (Fig. 5a-b). These range from  $< 100 \mu\text{m}$  to  $> 1$   
321 mm in size, with similar sizes of fluffs tending to be spatially correlated, defining  
322 discrete domains. The domains are sometimes lense-like indicating a probable  
323 sedimentary origin; this might suggest some periodicity to sediment input. Though  
324 some fluffs have a high aspect ratio and tapering edges, most are sub-spherical. In all  
325 sections, regardless of orientation relative to bedding, these grains have generally

326 cloud-like, ‘fluffy’ morphologies. They frequently have inclusions of wisps of silica  
327 and isolated, euhedral-subhedral opaque crystals, which together constitute < 20 % of  
328 the grain (Fig. 5a-b). We interpret these fluffy grains as carbon-impregnated silicified  
329 volcanic ash (cf. Lowe, 1999; Brasier et al., 2006). Their considerable interparticle  
330 (~30%) and intraparticle porosities supports this hypothesis, as does their multiclastic  
331 composite constructions (Fig. 5b), suggesting either in-air clotting of multiple ash  
332 grains when moistened or submarine or water-surface moistening and coagulation.  
333 Elemental mapping shows elevated concentrations of aluminium and potassium  
334 within the fluffy grains (Supp. Fig. 3a), also consistent with an origin as volcanic ash  
335 (Nakagawa and Ohba, 2003). Furthermore, these grains strongly resemble silicified  
336 volcanoclastics described from other members of both the Warrawoona Group and the  
337 time-equivalent Onverwacht Group of South Africa (Lowe and Knauth, 1977, 1978;  
338 Walsh, 2004; Walsh and Lowe, 1999). Raman micro-spectroscopy confirms the  
339 carbonaceous composition of these fluffy grains (Fig. 5c) and shows that carbon  
340 impregnation occurred prior to the maximum metamorphic or hydrothermal heating of  
341 these rocks in the early-mid Archean (Fig. 5d).

342

343 The second most common grain type, accounting for approximately 30% of most  
344 microgranular cherts, is the ‘flake’, which is a considerably darker, tapered, elongate  
345 grain (Fig. 6a-c). Most flakes are shallowly curved (Fig. 6c); since no way-up criteria  
346 are available in what we here interpret as a reworked sediment, no inference of  
347 concavity or convexity is implied. Flakes are largely restricted to the microgranular  
348 chert, suggesting either a transient formational mechanism, or small reservoir of  
349 material from which flaky grains can be drawn, preventing more widespread  
350 preservation. Flaky grains appear more densely carbonaceous than fluffy lobate

351 grains, and resemble either: i) ripped-up slivers and laminae of fine-grained sediment  
352 (cf. Schieber et al., 2012); ii) ripped-up chips of microbial mats (Noffke, 2010 and  
353 references therein); or iii) compressed fluffy grains. Where thin sections are cut  
354 perpendicular to the macroscopic banding, there is an obvious preferred orientation of  
355 elongate clasts, which is lost in thin sections cut parallel to macroscopic banding.  
356 These oriented flakes usually dominate granular layers alternately to the  
357 aforementioned fluffs (Fig. 4e). Raman micro-spectroscopy confirms the dominantly  
358 carbonaceous composition of flaky grains (Fig. 6a-b), and the carbon 'D' and 'G'  
359 peak intensities, position and shapes of these grains are identical to the  
360 aforementioned fluffy grains.

361

362 One explanation for the flaky grains is that they are compressed lobate fluffy grains.  
363 Rare, partially compressed fluffy grains are observed in silicified ashes at Chinaman  
364 Creek; these have tapering margins and jagged bifurcations and compaction is  
365 suggested by sutured contacts between adjacent grains (Fig. 6d). However, such  
366 compressed lobate clasts with sutured margins have not been observed in the  
367 microgranular chert. In addition, when the intra-grain features of known compressed  
368 lobate clasts (Walsh and Lowe, 1999) are compared to the flaky grains in the  
369 microgranular cherts, the two bear only superficial morphological resemblance. For  
370 example, inclusions are rare to absent in the flaky grains described herein, but are  
371 common in known compressed lobate clasts. Grain morphologies of the flaky grains  
372 suggest a more linear, fissile breakage than the irregular outlines of high aspect ratio  
373 lobate grains (Walsh and Lowe, 1999). Flaky grains also appear to lack the  
374 enrichment in aluminium and potassium observed in the fluffy grains (Supp. Fig. 3).  
375 Furthermore, the occasional occurrence of flaky grains immediately adjacent to



376 uncompressed fluffy grains within a sediment layer (Fig. 6b) discounts a mechanism  
377 whereby changes in silicification style may allow one layer to remain uncompressed  
378 (fluffy grains) while the next layer becomes compressed (flaky grains).

379

380 An alternative mechanism for the generation of flakes can be found in modern flume  
381 experiments on very fine-grained sediments. These suggest that above a certain flow  
382 velocity threshold, flake-like fragments of sediment can be removed from water-  
383 saturated muds, and redeposited as flakes once flow velocity dissipates (Schieber et  
384 al., 2012). A similar mechanism could also explain flake genesis as fragments of  
385 microbial biofilms eroded off the edges of a larger parent mat (cf. Noffke et al., 2013  
386 fig. 17).

387

388 The final type of clast found in microgranular cherts is the laminated grain (Figs. 7-8,  
389 Supp. Fig. 2); these are much rarer, making up < 5 % of chert volume. Laminated  
390 grains can be separated into two categories: i) those which are inherently laminated,  
391 i.e. primary lamination (Fig. 7), and ii) those which show lamination resulting from  
392 secondary intrusion or dilation by silica, either from the matrix, or from later clear  
393 microcrystalline veins (Fig. 8). Both are described in more detail in section 4.2.2.1  
394 below.

395

#### 396 4.2.2. Laminated textures

397 Lamination occurs on a variety of scales. The microgranular cherts are  
398 laminated on the mm-scale and these laminae appear to be defined by the  
399 relative proportions of fluffy and flaky grains (Fig. 4e, Supp. Fig. 1). Laminae  
400 also occur within single, albeit rare, grains in the microgranular chert (Figs. 7-8,

401 Supp. Fig. 2), mostly being defined by the relative proportions of carbon and  
402 silica at the tens of microns scale (see section 4.2.2.1 below). Finally,  
403 carbonaceous laminae can persist across entire thin sections cut perpendicular to  
404 bedding, and can be stacked together vertically for several millimetres in chert  
405 zones adjacent to microgranular zones (Fig. 9; see section 4.2.2.2 below). Some  
406 of these laminae appear filament-like (cf. Noffke, 2009, 2010) rather than being  
407 layered planar surfaces.

408

#### 409 *4.2.2.1. Laminated grains*

410 *Primary laminated grains:* Primary laminated grains within the microgranular chert  
411 feature gradation between their alternating siliceous and carbonaceous laminae. They  
412 display an ordered repetition of lamination that could indicate either an environmental  
413 or biological periodic oscillation acting on the precursor sediment. Dark carbonaceous  
414 and pale siliceous laminae occur sequentially on the scale of tens of microns, with  
415 neither predominating (Fig. 7a-d). Raman micro-spectroscopy confirms that the dark  
416 bands are indeed carbonaceous (Fig. 10b) while NanoSIMS ion mapping shows that  
417 carbon, nitrogen and sometimes sulfur co-occur in enhanced concentration in dark  
418 laminae (Fig. 11). The Raman spectra (Fig. 10c) show that the thermal maturity of the  
419 carbon is consistent with an early Archean age of deposition. The Raman spectra are  
420 qualitatively near identical to those described previously from the stratiform Apex  
421 chert (Sforna et al., 2014). It is not possible to compare our spectra quantitatively with  
422 those of Sforna et al (2014) because we used a different laser wavelength (785 nm as  
423 opposed to 514 nm) for excitation of the sample, which has been shown to induce a  
424 shift in the carbon D peak position (Pocsik et al., 1998). Hence, our Raman data only  
425 confirm that the carbon in the laminae experienced a similar degree of heating to

426 previously reported carbon, hence is likely an early phase, but cannot distinguish  
427 whether the carbon was sourced from a biological or other (e.g., deep hydrothermal)  
428 reservoir.

429

430 One character common to all of the grains herein interpreted as primarily laminated is  
431 that their laminae are non-isopachous, with thickness varying significantly over tens  
432 of microns along bands (Fig. 7). Many laminated grains show a thickening of the  
433 carbonaceous material toward the ‘crests’ of individual gently undulose laminae (Fig.  
434 7). Some grains demonstrate particularly undulose and wrinkled laminations (Fig. 7)  
435 and/or the rolling-up of multiple laminations (Figs. 7b, 12). Siliceous bands exhibit a  
436 crystallisation texture that appears to be influenced by the adjacent carbonaceous  
437 material - for example, malformed growth of otherwise euhedral microquartz crystals,  
438 further suggesting that carbonaceous laminae were likely lithified in their non-  
439 isopachous form.

440

441 The three-dimensional morphology of carbonaceous material in laminated grains is  
442 highlighted by autofluorescence under confocal laser scanning microscopy (CLSM),  
443 which demonstrates a wrinkled planar form (Fig. 13; supplementary movie 1) i.e., it is  
444 neither filamentous nor tubular. Many of the thicker carbonaceous bands appear  
445 multi-laminar in both light microscopy (Fig. 7d) and CLSM (Fig. 13). The largest  
446 laminated grain examined by CLSM (from locality CC8 of Brasier et al., 2011)  
447 reveals the rolling up of the tapering ends of several laminae; in some cases, these  
448 almost roll over by 180° (Fig. 13c-d; supplementary movie 1). Rollups such as these  
449 have been proposed as evidence for an initial plasticity of structure and are commonly  
450 associated with ancient microbial mats (Tice & Lowe, 2004; Tice et al., 2011). Roll-

451 up formation is thought to begin with the erosion, by waves, tides, or other marine  
452 current movements, of the original mat. This releases fragments of the mat and leaves  
453 tear-up structures in the remaining microbial edifice (Westall et al., 2015). Current  
454 movements have a range of erosional outcomes, depending foremost on the  
455 morphology and geometry of the microbial mat (Tice et al., 2011). However, if an  
456 eroded mat fragment is glutinously bound by cohesive extra-cellular polymeric  
457 substance (EPS), current action will deform, but not structurally disintegrate, the  
458 fragment, reworking it into a roll-up (cf. Tice and Lowe, 2004).

459

460 *Secondary laminated grains:* Some laminated grains appear secondary in nature and  
461 more closely resemble fragmented flakes. Here, micro-quartz veins traverse and  
462 intrude the clast disconformably, resulting in sharp contrasts between laminae (Fig.  
463 8a-c). The repetitively alternating banding characteristic of primary fabrics is not  
464 present. If these laminae were syn-sedimentary or biologically mediated, a gradation  
465 between carbonaceous and siliceous layers would be expected, as observed in the  
466 previously described primary grains, and indicative of incremental growth of the  
467 precursor sediment and/or biological system. The lamina boundaries here, however,  
468 are distinct, signifying brittle breakage or intrusion. Where ‘gradation’ at the margins  
469 of laminae is noted, higher magnification observation reveals this to be a result of  
470 secondarily loosened darker grains from the adjacent carbonaceous material, likely  
471 through pervasive silicification enhancing fracture defects (Fig. 8c). The formational  
472 process for these laminated grains is best explained as the intrusion of siliceous fluids  
473 into planes of weakness or fracture in the precursor clast, forcing material apart. This  
474 process is supported by two further lines of evidence, in addition to the sharp laminar  
475 edges. Firstly, the lighter, siliceous bands do not always traverse the entirety of the

476 clast (i.e., there are not always continuous planes as would be expected for a  
477 microbial mat or regularly repeating sediment deposition) and are aligned randomly  
478 (i.e., they are not likely related to the primary laminated fabric of the microclastic  
479 chert). Secondly, the undulations of the carbonaceous laminae appear to be an  
480 intimately related response to forcing caused by intercalated growth of silica: there is  
481 a near perfect fit between now-separated flakes which leads to our interpretation of  
482 these fragments as having once been parts of a larger precursor grain (see particularly  
483 the central and lower major grains in Fig. 8a).

484

485 *Other laminated grains:* Some of the most finely laminated grains fit into neither of  
486 the aforementioned categories. In these cases, laminae are distinctly isopachous, have  
487 tapered edges, and can be discontinuous across a clast. In contrast to all other laminae,  
488 these are composed of very fine yellow and orange-brown grains, only visible at high  
489 magnification (Fig. 8d). Carbonaceous material is absent, and these grains show  
490 strong resemblance to tubular pumice or welded tuff (cf. Klug et al., 2002; Polacci,  
491 2005). Similar microstructures have also been observed in stratigraphically higher  
492 Apex Basalt (Matthewman, pers. comm.).

493

#### 494 4.2.2.2. Mesoscopic 'filament-like' laminations

495 At two localities (CC164 and CCT23 of Brasier et al., 2011), mesoscopic laminations  
496 occur in microcrystalline black-grey-white chert zones (we interpret this lithology as  
497 an end-member classification of microgranular chert, and it should not be confused  
498 with the black-grey-white banded cherts, which are independent) adjacent to  
499 microgranular zones. These comprise narrow filament-like textures with an  
500 undulating, wrinkled topography (Fig. 9b, dashed arrows). The laminae are stacked

501 vertically into packages varying from < 100  $\mu\text{m}$  to several millimetres thick.  
502 Individual filament-like laminae within each package are rather diffuse, but the best-  
503 preserved examples are about 10-20  $\mu\text{m}$  thick (Fig. 9d, dashed arrows). The lateral  
504 extent of these laminae can be as little as a few tens of micrometers or they may  
505 persist across an entire thin section (Fig. 9a) or hand specimen. Laminae are  
506 interspersed with microcrystalline silica and this overall texture is sometimes cross-  
507 cut by later micro-quartz veins. A number of aggregations of these laminae appear to  
508 entrain orientated detrital sediment grains (Figs. 9c-e, 14). Raman micro-spectroscopy  
509 confirms that the laminations are carbonaceous, that the carbon is not a modern  
510 contaminant, and that the trapped grains are quartz (Fig. 14). This is consistent with  
511 the trapping and binding of sediment grains by filamentous microbes and their  
512 associated exopolymeric substances, as observed in modern microbial mats, and  
513 several fossil examples of Archean age (e.g., Noffke et al., 2001, 2003, 2013; Westall  
514 et al., 2011).

515

#### 516 4.2.3. Spherulitic textures

517 The stratiform cherts are often punctuated by spherulitic features. For example,  
518 irregular shapes exhibited by some carbonaceous fluffy grains in clotted and  
519 microgranular cherts - shapes in marked contrast to the dominant sub-rounded, cloud-  
520 like morphologies of these grains - could result from their fragmentation by silica  
521 spherules. Fragmentary, homogeneous, and indistinct carbonaceous clasts are often  
522 present around lobate grain margins in zones of spherulitic alteration; these may  
523 signify the breakup products of larger lobate parent bodies by spherulitic silica  
524 growth. These spherulitic textures are often associated with elongate, sometimes  
525 partially-filamentous fragments of carbon, that resemble abiogenic pseudofossils

526 previously identified in the black chert veins below the stratiform chert (Brasier et al.  
527 2005, 2011).

528

## 529 **5. Discussion**

### 530 *5.1. Assessment of Biogenicity*

531 A number of the carbonaceous microstructures detailed above are comparable to  
532 features previously interpreted as biosignatures in Archean and Proterozoic rocks  
533 (e.g., Noffke et al., 2003, 2006, 2013; Tice and Lowe, 2004). Microstructures of  
534 particular interest identified through our appraisal of the stratiform Apex chert are: i)  
535 primary laminated grains; ii) filament-like laminae entraining sediment grains; iii)  
536 roll-up structures; and iv) flaky grains. Combined, these microstructures resemble a  
537 suite of microscopic microbially induced sedimentary structures (MISS) as defined by  
538 Noffke (2009, 2010), and described from both modern (e.g., Noffke et al., 2001) and  
539 ancient (e.g., Noffke et al., 2003) environments. Here we test the biogenicity of these  
540 Apex microstructures against a suite of biogenicity criteria (e.g., Schopf and Walter,  
541 1983; Buick, 1990; Brasier et al., 2004; Hofmann, 2004; Wacey, 2009; Noffke 2010).  
542 Some of these criteria are specific to MISS, whilst others are more generic, applicable  
543 to any putative Precambrian biogenic structure. The ‘fluffy’ grains, spherulitic  
544 microstructures and secondarily laminated grains will not form a substantial part of  
545 our discussion because there is little or no suggestion from their morphology that they  
546 might be biogenic. They are, however, useful as comparative material in our  
547 discussion of the putatively biogenic microstructures.

548

549 i) *Biogenic structures must occur in rocks of both known provenance and of*  
550 *demonstrable Archean age. Furthermore, the structures must be a part of, and*  
551 *syngenetic with, the primary fabric of the host rock.*

552 The stratiform 'Apex chert' has been extensively mapped from the kilometre down to  
553 the micrometre scale, and all samples can be relocated using their GPS coordinates. It  
554 is a sedimentary unit, cropping out for several hundred metres along strike, and is  
555 located within a well-constrained stratigraphic column, with radiometric dates from  
556 both above and below the unit (e.g., Van Kranendonk, 2006). The microstructures of  
557 interest occur either within grains and clasts that have been eroded from older units  
558 and then incorporated into the microgranular chert (e.g. primary laminated grains), or  
559 are part of the primary fabric of the rock (e.g. mesoscopic 'filamentous' laminations).  
560 Raman micro-spectroscopy shows that all carbon has a thermal maturity consistent  
561 with an early Archean age (i.e. emplacement prior to peak metamorphic/hydrothermal  
562 temperatures experienced by the Apex succession sometime prior to 3 Ga; Van  
563 Kranendonk, 2006; Sforza et al., 2014) thus cannot be a more modern contaminant.

564

565 ii) *Biogenic structures should not be found in metastable mineral phases, void-*  
566 *filling cements or veins.*

567 None of the microstructures of interest are found in such late-stage or metastable  
568 mineral phases; they occur either in clasts of micro-quartz that were lithified, eroded  
569 and reworked prior to the final lithification of the stratiform chert (e.g., Fig. 5a), or are  
570 found in primary laminated chert (e.g., Fig. 9). In contrast, spherulitic textures  
571 forming filamentous and other carbonaceous fragments are found in void filling  
572 cements and cross-cutting veins. These are demonstrably crystal-edge effects, usually  
573 resulting from the growth of silica spherules. The carbon in such later phases has been



574 redistributed to such an extent that its morphology cannot be used to determine its  
575 origin (cf. Pinti et al., 2009; Wacey et al., 2015).

576

577 *iii) Fossil MISS must occur in sedimentary rocks having undergone only low grades*  
578 *of metamorphism.*

579 This criterion is met by the sedimentary sequences in the Marble Bar greenstone belt,

580 which have not experienced more than lower greenschist facies regional

581 metamorphism (Hickman and Lipple, 1978; Van Kranendonk, 2006). More

582 specifically, a recent Raman study at the Chinaman Creek locality estimated that the

583 maximum temperatures experienced by these rocks were between 265°C and 360°C,

584 which may represent the peak temperature of regional metamorphism and of

585 hydrothermal fluids respectively (Sforna et al., 2014). Multiple potential biosignatures

586 have been reported previously from similarly silicified sediments of the Pilbara craton

587 (e.g., Sugitani et al., 2010, Wacey et al., 2011; Noffke et al., 2013) indicating that

588 MISS could also be preserved within the stratiform Apex chert.

589

590 *iv) The geological context of the lithology should be plausible for life; ideally, the*

591 *lithology should indicate a transgressive depositional phase, since this is the*

592 *environment in which modern MISS develop.*

593 The stratiform Apex chert is likely to represent a quiescent marine environment in

594 which low-density particles can settle out of suspension, i.e. lobate ash clasts in layers

595 and very fine clay or fragments of microbial mat. Water temperatures would be within

596 the range in which (hyper)thermophilic life could flourish; geochemical evidence,

597 including relatively high concentrations of barium in the stratiform chert and small

598 positive europium anomalies, indicate low-temperature (100-150°C) hydrothermal

599 venting (Kato and Nakamura, 2003). In addition, the input of volcanic detritus may  
600 have provided essential elements for life, thus the environment appears habitable.  
601 However, further high-resolution sedimentary logging is required to determine  
602 whether a transgression can be identified within the stratiform chert unit. This work  
603 should also search for evidence of macroscopic MISS that are as yet unreported from  
604 this unit.

605

606 v) *MISS are predominantly preserved in fine quartz-rich sediments in a moderately*  
607 *reworked hydraulic setting.*

608 The stratiform Apex chert partially meets this criterion, especially for the  
609 microgranular cherts (e.g. localities CC8 and CC117 of Brasier et al., 2011), which  
610 are interpreted as reworked sediments. The Apex stratiform rocks now have an almost  
611 uniform quartz composition. However, since widespread silicification has altered their  
612 original composition, this implies little regarding deposition in a siliciclastic system.  
613 Initial grain size is also difficult to determine, having been extensively modified by  
614 various silicification events, though the rare trapped grains in the filament-like lamina  
615 (Fig. 9) are comparable in size to those found in other Archean and Proterozoic MISS  
616 (e.g., Noffke et al., 2003, 2006, 2013).

617

618 vi) *Structures should fit into a plausible evolutionary context and would ideally*  
619 *exhibit community behavior.*

620 Microbial mats are demonstrably one of the most ancient and enduring biological  
621 communities, with evidence of microbial mat/sediment interaction reported from  
622 rocks of similar age to the Apex chert in both the Pilbara and Barberton regions (e.g.,  
623 Tice and Lowe, 2004; Allwood et al., 2006; Westall et al., 2001, 2006, 2015; Noffke

624 et al., 2013). An interpretation of the laminated and filamentous Apex microstructures  
625 as MISS fits within this evolutionary context and, by analogy to modern ecosystems,  
626 implies community behavior of a microbial mat (Noffke, 2008; Schieber et al., 2007).  
627 We do not interpret any of the microstructures as microfossils, though we raise the  
628 possibility that the filament-like laminae entraining sediment grains may be the  
629 diffuse remnants of mat-building filamentous microorganisms (cf. Noffke et al.,  
630 2003). Further detailed geochemical and morphological research must be conducted  
631 into these, however, before such an interpretation can be substantiated.

632

633 *vii) Laminated MISS should be wavy or wrinkled, with several orders of curvature*  
634 *i.e. should not be uniform crusts, which are usually precipitative. Thickening of*  
635 *carbonaceous laminae towards the crests of laminae would provide additional*  
636 *evidence of biology (cf. Pope and Grotzinger, 2000).*

637 Both the primary laminated grains and the filament-like laminae are often wavy and  
638 wrinkled at the mesoscopic and microscopic scale (Figs. 7, 9, 12-14). In addition,  
639 some of the laminae within larger grains in the microgranular chert thicken toward  
640 undulose crests. In contrast, such features are absent from the secondarily-laminated  
641 carbonaceous grains (Fig. 8a-c) and from isopachous microstructures here interpreted  
642 as tubular pumice (Fig. 8d). Within the metalliferous cherts and black, grey and white  
643 banded cherts (Fig. 4g-j), there are laminations that are continuous across thin  
644 sections, often on the same sub-millimetre scale as the laminae described in the  
645 microclastic cherts. These laminae are, however, isopachous and show nothing of the  
646 multiple orders of curvature required for a biological interpretation. Hence, they are  
647 interpreted as precipitative crusts growing sequentially over other fabric elements in  
648 the cherts.

649

650 ix) *MISS must be shown to preserve textures that either represent, have been caused*  
651 *by, or are related to, biofilms or microbial mats.*

652 In addition to the wavy and wrinkled laminae described above, some of the putative  
653 MISS contain grains around which the filament-like laminae wrap. In modern  
654 biofilms, microbial mats and stromatolites, such microstructures form via the trapping  
655 and binding of sediment grains by ‘sticky’ filamentous microorganisms and their  
656 associated extra-cellular polymeric substances (EPS; e.g., Reid et al., 2000).

657 Similarly, rolled-up microstructures in modern settings are cited as evidence of an  
658 initial plasticity, hence cohesiveness in the sediment, and ancient examples have been  
659 interpreted as resulting from the interaction of erosive forces, such as currents, with a  
660 microbial mat (e.g., Tice and Lowe, 2004). We have identified putative roll-ups in the  
661 microclastic cherts (CC8 and CCT27), though they are of smaller size than well-  
662 accepted Archean examples (Tice and Lowe, 2004). Additionally, a plausible  
663 interpretation of the flake-like grains reported here is that they are micro-scale  
664 analogues of microbial mat chips commonly found in modern MISS assemblages,  
665 also formed when water agitation (driven by tides or storms, for example) tears small  
666 pockets of semi-cohesive material from their parent microbial mat (Gerdes and  
667 Krumbein, 1987, Tice et al., 2011).

668

669 x) *MISS should possess geochemical signals indicative of biology*

670 Both laser Raman micro-spectroscopy and NanoSIMS data show that the laminated  
671 microstructures are carbonaceous. The Raman spectra are consistent with previous  
672 data from ancient biological material within greenschist facies rocks (e.g., Tice et al.,  
673 2004) and with previous data from the Apex stratiform chert (Sforna et al., 2014).

674 Raman data cannot prove the biogenicity of organic material, because similar spectral  
675 features can be obtained from non-biological organic matter (Pasteris and Wopenka,  
676 2003). NanoSIMS data show the co-occurrence of carbon, nitrogen and sometimes  
677 sulfur within the dark laminae of primary laminated grains. These data are not  
678 quantitative, but they do demonstrate that three of the elements integral to life occur  
679 in elevated concentrations in zones that also have a microbial mat-like morphology.  
680 Similar correlations of microbial morphology with biologically significant elements  
681 have been demonstrated within modern and ancient stromatolites (Wacey, 2010) and  
682 *bona fide* Precambrian microfossils (Oehler et al., 2006; Wacey et al., 2011).

683

#### 684 *5.2. Summary of potential biogenicity*

685 Of the range of textures exhibited by the stratiform cherts, we find that it is the  
686 microgranular cherts that hold the most promise for the retention of biosignatures.  
687 Primary laminated grains, filament-like wrinkle structures, roll-ups, and flaky grains  
688 may all plausibly be interpreted as remnants of a microbial mat community. The  
689 texture of the microclastic cherts suggests a shallow, quiescent environment of  
690 deposition, in which weak but persistent currents facilitated the orientation and  
691 imbrication of elongate clasts. There is a prominent volcanoclastic component to the  
692 microgranular cherts, and evidence of a proximal silicic source comes in the form of  
693 interbedded silicified ashes and other volcanoclastics that constitute much of the  
694 stratiform stratigraphy at Chinaman Creek (Kato and Nakamura, 2003; Brasier et al.,  
695 2011). It may be that this volcanic input provided elements, particularly metals,  
696 significant to the emergence of life in this habitat (cf. Barras, 2012; Van Kranendonk,  
697 2006). However, the lack of evidence for macroscopic MISS or definitive  
698 microfossils in this unit urges a note of caution. In addition, the environment of

699 deposition and hydrothermal style of silicification (e.g., Kato and Nakamura, 2003),  
700 overprinting some of the primary sedimentary features, is rather different from that of  
701 traditional siliciclastic settings in which MISS are well understood (cf. Noffke, 2010),  
702 and for which the criteria outlined above were primarily designed.

703

704 Other lithologies in the stratiform sequence at Chinaman Creek, namely banded grey  
705 and white and metalliferous cherts, clotted carbonaceous cherts and silicified  
706 volcanoclastics, are more difficult to decode. They have not yet provided putative  
707 biogenic structures, though they are important for diagnosing environmental and  
708 redox conditions during Apex time. The clotted cherts, in particular, possess textures  
709 generated from spherulitic silica growth, during which microfossil-like artifacts  
710 developed around the margins of crystals. The carbon in these textures may ultimately  
711 have a biogenic origin, but abiogenic sources, for example through hydrothermally-  
712 mediated processes (e.g., Fischer-Tropsch synthesis), remain an equally plausible  
713 explanation for this carbon (cf. Brasier et al., 2005).

714

715 This study adds to the growing evidence for a diversity of primitive life in the early  
716 Archaean era, and provides a detailed assessment of carbonaceous microstructures  
717 within the stratiform Apex chert at Chinaman Creek, a lithology that has been only  
718 very briefly described in previous work (e.g., Brasier et al., 2011; Sforza et al., 2014).  
719 Our data support the hypothesis that shallow-water environments, together with input  
720 from volcanic and hydrothermal sources, were likely pivotal niches occupied by  
721 simple, prokaryotic mat-forming organisms. The MISS-type structures described  
722 herein are found in relatively close proximity to penecontemporaneous hydrothermal  
723 fabrics, yet have no apparent genetic association with these higher-temperature

724 events; thus their interpretation as biological signals is more probable than an abiotic  
725 origin.

726

## 727 **5. Conclusions**

728 The stratiform 'Apex chert' at Chinaman Creek is a varied and previously  
729 understudied lithological suite, which we have divided into five dominant components  
730 based on petrographic observations: i) carbonaceous laminated microgranular chert;  
731 ii) laminated black, grey and white chert; iii) metalliferous (Fe-rich) chert; iv) clotted  
732 carbonaceous chert; and v) silicified volcanoclastics. The protoliths of all stratiform  
733 chert rocks have been pervasively and ubiquitously silicified, and a dominant  
734 component of this silicification was low temperature (100-150°C) hydrothermal fluids  
735 (Kato and Nakamura, 2003).

736

737 The macroscopic sedimentary textures of the stratiform Apex cherts, which have  
738 dominant silt-grade grains in most localities studied, and occasional imbrication of  
739 semi-lithified chert fragments, hints at a shallow marine depositional environment.  
740 The poor lateral continuity of individual chert layers argues against deep marine  
741 pelagic settling of sediment. The depositional environment is herein interpreted to be  
742 a protected shallow marine environment, where weak currents are intermittently  
743 present to sort and orient clasts. Inter-bedded metalliferous cherts, either ferruginous  
744 or jaspilitic, may signify a change in redox state or temperature, most likely linked to  
745 the penecontemporaneous hydrothermal venting and the intrusive black chert veins  
746 (Kato and Nakamura, 2003; Brasier et al., 2005, 2011). Volcanic components, largely  
747 ashfall, are prevalent throughout the stratiform sequence, consistent with a shallow-  
748 marine environment situated adjacent to active volcanoes. This is similar to the

749 environmental setting described for other units from both the Pilbara and Barberton  
750 regions containing putative biological remains of approximately equivalent age (e.g.,  
751 Kitty's Gap Chert and Josefsdal Chert; Westall et al., 2006, 2011, 2015). The  
752 stratiform Apex chert depositional environment is not compatible with the deeper  
753 marine regime suggested for some Archean cherts (Lowe, 1984; Paris et al. 1985).

754

755 Carbonaceous material is especially abundant in the microgranular and clotted cherts,  
756 and is particularly concentrated in grains. Carbonaceous textures interpreted to have a  
757 biological component are present in the microgranular cherts as four morphotypes.

758 Firstly, there are carbonaceous laminated grains that pass multiple geological,  
759 morphological and geochemical criteria for biogenicity and antiquity. Secondly, more  
760 pervasive carbonaceous filament-like laminations are present, which entrain sediment  
761 grains and closely resemble microscopic MISS. Thirdly, we report elongate, flake like  
762 carbonaceous grains, which potentially represent eroded, 'ripped-up' fragments of a  
763 microbial mat (cf. Noffke, 2009). We caution that further work is required on the  
764 flake-like grains to fully discount an origin from a purely sedimentary protolith (cf.  
765 Schieber et al., 2012). Finally, rare roll-up structures are present, both as part of larger  
766 laminated grains, and as isolated features in the matrix of microclastic cherts. These  
767 are interpreted as current-eroded and plastically reshaped microbial mat fragments,  
768 similar to those described from the ~3.4 Ga Buck Reef Chert (Tice and Lowe, 2004;  
769 Tice et al. 2011).

770

771 CLSM is here shown to be an effective technique for the imaging of carbonaceous  
772 microstructures in these Archean cherts; the autofluorescence of carbonaceous  
773 material produces sequential tomograms through the depth of all thin sections



774 investigated herein. This technique enables the generation of sub-micrometer-scale  
775 spatial resolution, three-dimensional renderings of features of biogenic interest, and  
776 here strengthens the morphological evidence for the biogenicity of some of the  
777 described laminated carbonaceous textures. NanoSIMS and laser Raman provide  
778 evidence for the concentration of life-significant elements in microstructures that  
779 closely resemble the morphology of microbialites. An encouraging combination of  
780 both morphology and chemistry pertinent to life in the laminated microstructures of  
781 the stratiform cherts, a lithology representing a geologically plausible environment for  
782 microbial life, suggests that such features have a biological origin. Although no  
783 compelling evidence for life has been found in the underlying heavily studied black  
784 chert veins (Brasier et al., 2002, 2005, 2006, 2011, 2015; Wacey et al., 2015; Bower  
785 et al., 2016), evidence of an early Archean biological community may yet be present  
786 at Chinaman Creek.

787

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806

807

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### 1132 **Figure Captions**

1133

1134 **Figure 1.** Location of the Chinaman Creek study area. a) Overview of the geology of  
1135 the East Pilbara Terrane, showing a series of domed granitoid complexes intruding  
1136 and overlain by volcano-sedimentary rocks of the Pilbara Supergroup. The Chinaman  
1137 Creek locality (red box) is found within the Marble Bar greenstone belt around 5 km  
1138 west of the town of Marble Bar. b) Geographical context of the East Pilbara Terrane  
1139 in Western Australia. Modified from Hickman (2008) and Brasier et al., 2011.

1140

1141 **Figure 2.** Field photograph looking southwest showing the south block of the ‘Apex  
1142 chert’. The stratiform chert (outlined in yellow) outcrops along the northwest-  
1143 southeast trending ridge, while hydrothermal black chert veins (arrowed in red and  
1144 labeled following the convention of Brasier et al., 2005, 2011) cut up through the  
1145 underlying basalt often inter-fingering with (but not passing entirely through) the  
1146 stratiform chert. Person for scale.

1147

1148 **Figure 3.** Geological map of the ‘Apex chert’ in the Chinaman Creek area. The area  
1149 consists of three structural blocks, north, central, and south, defined by growth faults.  
1150 The stratiform chert (unit 4 of Brasier et al., 2005, 2011) is the focus of this study and  
1151 outcrops continuously in both the south and north blocks. Black chert veins cut up  
1152 through the underlying basalt and underplate and interfinger with the stratiform chert.  
1153 The N1 vein houses the controversial ‘microfossil’ site of Schopf (1993). Locations of  
1154 samples analysed in this study are numbered. Modified from Brasier et al. (2011).

1155

1156 **Figure 4.** Scans of geological thin sections, each accompanied by plane polarized  
1157 light photomicrographs of sub-portions of the thin section, showing typical fabrics  
1158 found within the stratiform ‘Apex chert’. a-b) Silicified ash. c-d) Clotted  
1159 carbonaceous chert. e-f) Banded microgranular chert (red and white arrows denote  
1160 alternating bands dominated by differing grain types), see also Supplementary Figures  
1161 1 and 2. g-h) Metalliferous (Fe-rich) chert. i-j) Banded black-grey-white chert.

1162

1163 **Figure 5.** Lobate fluffy grains from the stratiform chert. a-b) Typical lobate fluffy  
1164 grains within microgranular chert with inclusions of wisps of chert and opaque  
1165 crystals. Many large lobate grains (e.g., b) appear to be composites of multiple



1166 smaller grains, consistent with an origin as carbon impregnated, clotted volcanic ash  
1167 grains. c) Raman image from the edge of a fluffy grain showing its carbonaceous  
1168 composition (red) and the quartz matrix (green). d) Typical Raman spectrum from a  
1169 fluffy grain; note the well-developed carbon 'D' and 'G' peaks and small quartz peak.  
1170

1171 **Figure 6.** Flake-like grains from the stratiform chert. a-b) Flake-like grains from the  
1172 microgranular chert with accompanying Raman maps illustrating their carbonaceous  
1173 (red) plus minor quartz (green) composition. Raman maps are from areas indicated by  
1174 the yellow arrows and were constructed using the integrated intensities of the  $\sim 1600$   
1175  $\text{cm}^{-1}$  carbon 'G' Raman band and the  $\sim 465 \text{ cm}^{-1}$  quartz Raman band respectively. c)  
1176 Elongate, curved, tapering flakes within microgranular chert. This is the dominant  
1177 flake morphology in these cherts. d) Compressed lobate fluffy grains in silicified  
1178 volcanic ash, with compression indicated by suturing of grains (arrows); their  
1179 morphology superficially resembles flakes but their internal texture and chemistry  
1180 distinguishes them from true flakes found in the microgranular chert.

1181

1182 **Figure 7.** Primary laminated grains from the microgranular chert. a) Non-isopachous  
1183 laminated microstructure in a region of otherwise fluffy lobate grains, interpreted as a  
1184 reworked fragment of a larger laminated parent body. b-c) Non-isopachous, undulose  
1185 and faintly crinkled laminated grains; note thickening of carbonaceous lamina at ridge  
1186 crest (red arrow) and potential rolling up of upper thick carbonaceous lamina (yellow  
1187 arrow). d) Thick carbonaceous band within a laminated grain composed of multiple  
1188 finer carbonaceous laminae (arrow).

1189

1190 **Figure 8.** Non-primary laminated grains from the stratiform chert. a-b) Silica  
1191 intrusion into fluffy or flaky grains giving the impression of lamination. The silica  
1192 ‘laminae’ frequently do not traverse the whole grain and do not show the regular  
1193 repeating pattern of primary lamination. The carbonaceous portions of the grain are  
1194 often modified (e.g. pushed apart) by the intruding silica. c) Close up of a carbon-  
1195 silica boundary suggesting that carbonaceous material has been loosened by intruding  
1196 silica (arrow), leading to a false gradation between layers. d) Microstructure that  
1197 approximates tubular pumice, comprising isopachous laminae of tiny yellow-brown  
1198 altered volcanic glass grains without carbon.

1199

1200 **Figure 9.** Mesoscale filament-like laminations in the stratiform chert. a) Scan of  
1201 geological thin section (CC164) showing wrinkled, undulose laminae traversing the  
1202 entire thin section (upper half of image) adjacent to a microgranular chert zone (lower  
1203 portion of image). b) Plane polarized light image from the thin section shown in (a)  
1204 showing dark filament-like laminae (e.g., dashed arrows), often stacked vertically into  
1205 bundles several hundred micrometers thick. c) Image of the same thin section taken  
1206 under crossed-polars showing quartz grains (e.g. arrows) entrained within some of the  
1207 dark laminae. d) Higher magnification image from CC164 showing at least three  
1208 quartz grains (solid arrows) entrained within filament-like laminae (dashed arrows),  
1209 having their long axes parallel or at a shallow angle to the laminae. e) Particularly  
1210 dense carbonaceous laminae within sample CCT23 wrapping around a number of  
1211 quartz grains (examples arrowed). Again their long axes show similar alignment sub-  
1212 parallel to the trend of the laminae.

1213

1214 **Figure 10.** Mineralogy of laminated grains. a) A typical non-isopachous laminated  
1215 grain from the microgranular chert (sample CC8). b) A two-colour overlay Raman  
1216 map from area indicated in (a) showing the carbonaceous (red) and quartz (green)  
1217 composition of the alternating bands. c) A typical Raman spectrum from a  
1218 carbonaceous band illustrating the position and shapes of the carbon ‘D’ and ‘G’  
1219 peaks, plus small quartz peak. The carbon peak shapes are near identical to those from  
1220 the fluffy and flaky grains (e.g., Fig. 5d).

1221

1222 **Figure 11.** Geochemistry of laminated grains. a) A typical non-isopachous laminated  
1223 grain from the microgranular chert (sample CC8). b) NanoSIMS ion maps of carbon  
1224 ( $^{12}\text{C}^-$ ), nitrogen ( $^{12}\text{C}^{14}\text{N}^-$ ) and sulfur ( $^{32}\text{S}^-$ ), showing a positive correlation of carbon  
1225 and nitrogen, with relatively higher concentrations in the dark laminae (dashed lines  
1226 outline main dark laminae in analysis area). Sulfur correlates in part with carbon and  
1227 nitrogen, but we cannot rule out that it is also present in small mineral grains such as  
1228 pyrite. Brighter colours indicate relatively higher concentrations of each ion.

1229

1230 **Figure 12.** (a-c) Examples of putative roll-up microstructures within the  
1231 microgranular chert. In each case, the arrow indicates potential rolling up of thick  
1232 carbonaceous laminae.

1233

1234 **Figure 13.** Confocal laser scanning microscopy (CLSM) of primary laminated grains  
1235 from the microgranular chert. a) Entire confocal dataset taken from a laminated grain  
1236 in sample CC8-H2. The highly fluorescent spot to the lower centre is likely surface  
1237 contamination. b) Three-dimensional rendering from the dataset in a), oriented to  
1238 show the planar nature of the fluorescing carbonaceous laminations. These weakly

1239 undulate across the grain. c) Entire confocal dataset from a laminated grain in sample  
1240 CC8 displayed as a three-dimensional image, which again demonstrates a clear planar  
1241 character for the fluorescing carbonaceous laminae. d) Three-dimensional rendering  
1242 of the upper right portion of (c), highlighting the rolling up of laminae which  
1243 autofluoresce. These roll-ups (e.g., arrows) almost completely overturn reflexively;  
1244 roll-up is seen across at least seven of the upper laminae, and potentially in two  
1245 further lower laminae, thus is a common feature within this clast.

1246

1247 **Figure 14.** Chemistry of ‘mesoscopic’ laminations and entrained grains. a, c) Two  
1248 examples of entrained, orientated grains within dark filament-like laminae from  
1249 sample CCT23. b, d) Two colour overlay Raman maps from the areas indicated by the  
1250 blue boxes in (a) and (c) respectively, showing the carbonaceous content of the dark  
1251 laminae (red) and the quartz composition of the entrained grains (green). e) Typical  
1252 Raman spectrum from the dark laminae, exhibiting near identical carbon ‘D’ and ‘G’  
1253 bands to those shown by other primary carbonaceous microstructures in this unit (e.g.,  
1254 Figs. 5d and 10c). The rather diffuse nature of the carbonaceous filaments is shown by  
1255 the relatively strong Raman quartz bands indicating the filaments are now a mixture  
1256 of carbon and silica.

1257

1258 **Figure S1.** Textural characteristics of microclastic cherts at Chinaman Creek. a)  
1259 Microclastic chert (sample CC8) is unequivocally layered: in this scan of part of a thin  
1260 section, alternating darker and lighter coloured bands can be observed, which indicate  
1261 layers with a predominance of lobate fluffy ash clasts and elongate ‘rip-up’ flaky  
1262 clasts, respectively. b) A second microclastic chert (sample CCT27) also consists of  
1263 darker ash grains and lighter layers dominated by silica and flaky clasts, though here

1264 the banding is less distinct. There is a general coarsening-upward trend in grain size  
1265 here. c) Microcrystalline and megaquartz veins regularly cut across thin sections but  
1266 are rarely parallel to bedding. d) General texture of the microcrystalline cherts,  
1267 showing a relatively well-sorted, clast-supported texture. Scale bar is 5 mm in (a) and  
1268 (b) and 500  $\mu\text{m}$  in (c) and (d).

1269

1270 **Figure S2.** Variations of microclastic chert at Chinaman Creek. a) Slide scan of  
1271 microclastic chert (sample CC43), which is rich in orientated pale clasts, some of  
1272 which are banded. (b-c) Examples of banded clasts from sample CC43. Scale bar  
1273 equals 3 mm in (a) and 500  $\mu\text{m}$  in (b-c).

1274

1275 **Figure S3.** Elemental analysis of the microclastic chert (sample CC8). a)  
1276 Photomicrograph and elemental maps, showing enrichment of aluminium and  
1277 potassium within a fluffy lobate clast, supporting an interpretation that this clast has a  
1278 volcanic ash precursor. b) Photomicrograph and elemental maps (Al, C and O) from a  
1279 flaky clast; this lacks enrichment in aluminium (or potassium, not shown) and  
1280 possesses elevated levels of carbon.

1281

1282 **Movie S1.** Reconstruction of carbonaceous laminated grains from confocal laser  
1283 scanning microscopy data.

1284