Hickman-Lewis, K., Garwood, R. J., Brasier, M., Goral, T., Jiang, H., McLoughlin, N., & Wacey, D. (2016). Carbonaceous microstructures from sedimentary laminated chert within the 3.46 Ga Apex Basalt, Chinaman Creek locality, Pilbara, Western Australia. *Precambrian Research*, *278*, 161-178. https://doi.org/10.1016/j.precamres.2016.03.013

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

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Abstract

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

Herein, we document and assess the biogenicity of carbonaceous microstructures present in the lowermost of the stratiform chert units (informally known as the 'Apex chert'), at the Chinaman Creek locality in the Marble Bar greenstone belt, Pilbara Craton, Western Australia. Carbonaceous material mostly occurs within clotted grey-black cherts and microgranular 'grainstone-like' cherts within the stratiform unit, the latter being the major focus of this study. In the clotted cherts, carbon occurs as lobate, fluffy grains, rare compressed flakes, and as a grain boundary phase around spherulitic silica. There is no morphological evidence to support the biogenicity of these microstructures. In contrast, the microgranular chert contains fluffy and flaky carbonaceous grains, plus laminated grains comprising multiple non-isopachous wrinkled carbonaceous laminae, with noted thickening towards some ridge crests, as determined by confocal laser scanning microscopy. Roll-up structures provide evidence of an initial plasticity, interpreted to have formed via the tearing-up and current-induced plastic deformation of microbial mat fragments. Geochemical mapping, using laser Raman micro-spectroscopy and NanoSIMS, respectively demonstrates the antiquity of the carbon, and reveals a close correlation between carbon, nitrogen and sometimes sulfur, concentrated within dark brown to black laminae. Adjacent to microgranular zones are zones of more persistent carbonaceous, undulose, filament-like laminae that entrain relict sediment grains. These microstructures are directly comparable to a sub-type of microbially induced sedimentary structure (MISS), widely reported from younger siliciclastic sediments colonized by microbial biofilms.

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

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

1. Introduction

The 3.46 Ga Apex Basalt in the Marble Bar greenstone belt of the Pilbara Craton, Western Australia, has long been a focus for the investigation of early Earth environments and potential microbial life. It is particularly notable for the presence of carbonaceous filamentous microstructures in hydrothermal black chert veins at the Chinaman Creek locality, interpreted as representing eleven species of fossilized prokaryotes (Schopf and Packer, 1987; Schopf, 1992, 1993, 1999). However, the biogenicity of these 'microfossils' has been highly debated (e.g., Brasier et al., 2002, 2005, 2011; Schopf et al., 2002, 2007, 2010; Schopf and Kudryavtsev, 2009, 2012, 2013; Marshall et al., 2011; Ollcott Marshall and Marshall, 2013). Recent morphological and geochemical analyses at a high spatial resolution have shown the filaments to be mineral artifacts comprising chains of phyllosilicate crystals that later adsorbed carbon during fluid flow within an active hydrothermal system (Brasier et al., 2015; Wacey et al., 2015), or carbon-filled cracks (Bower et al., 2016). Biogenic stromatolitic clasts have also been postulated in the Apex hydrothermal chert veins (Schopf, 1993), though these were subsequently reappraised as isopachous, abiogenic, stromatoloidal internal cements, occurring in a later stage chert fabric (fabric B2 of Brasier et al., 2005).

Despite the controversy over the presence of life in the Apex hydrothermal chert veins, the Apex Basalt remains a promising rock unit to investigate early life on Earth. It is well-preserved, having undergone metamorphism to no greater extent than prehnite-pumpellyite to lower greenschist facies (Hickman and Lipple, 1978; Van Kranendonk et al., 2007). It also contains at least five stratiform chert units (Kato and Nakamura, 2003; Van Kranendonk, 2006); these are concordant, bedding parallel units that are often internally laminated and preserve sedimentary structures (Kato and Nakamura, 2003; Brasier et al., 2011). Hence, these may provide evidence of sedimentary environments (and their associated biotas) reflecting quiescent periods in an otherwise volcanic 'Apex time'. In some recent studies it has proven effective to look in volcanically influenced terranes for signs of early life (e.g., Walsh, 2004; Westall et al., 2006, 2015), since these provide many of the minor elements essential for life (e.g. Cu, Co, Ni and Fe; Barras, 2012). Volcanic lithologies have shown the capability to foster modern extremophilic life; shortly following their eruption, bacterial communities are able to benefit from the diverse, often metallic, elements present (Cockell et al., 2009; Kelly et al., 2011, 2014).

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

2. Methods

2.1. Field mapping and petrographic analysis

Field mapping in the Marble Bar greenstone belt was undertaken by us as part of a wider programme with the Geological Survey of Western Australia, supplemented by a detailed programme of mapping and sampling of the Apex Basalt across an area of around 12 km^2. Multiple samples were collected from the Chinaman Creek locality between 1999 and 2006, located by means of satellite images and Global Positioning Systems (GPS). For the stratiform 'Apex chert', samples were collected across the full 1.5 km of available outcrop, encompassing each of the north, central and south blocks described in Brasier et al. (2005, 2011). Optical petrography and fabric mapping was performed in the Department of Earth Sciences imaging laboratory, Oxford University, using standard 30 µm and 100 µm petrographic thin sections. Thin sections were examined under bright-field, polarized transmitted, and incident (reflected) light using Nikon Optiophot-2 (biological) and Optiophot-pol (polarizing) microscopes. Images were obtained using a single-chip CCD camera, providing live images in full RGB colour, and processed using AcQuis and Auto-Montage image capturing software.

2.2. Confocal Laser Scanning Microscopy (CLSM)

Confocal images were acquired for numerous features observed in thin-sections using a Nikon A1-Si laser-scanning confocal microscope using either a 20x or 40x oil-immersion objective (numerical apertures of 0.7 or 1.3, respectively). Images were recorded with pixel dimensions between 0.31 and 0.61 µm. Autofluorescence of the specimens was excited with the following laser lines: 405-nm line of 100 mW cube laser (Coherent Inc., USA, http://www.coherent.com), 488-nm line of 50 mW sapphire laser (Coherent Inc., USA), 561-nm line of 50 mW sapphire laser (Coherent Inc., USA) and 640-nm line of 40 mW cube laser (Coherent Inc., USA). Autofluorescence signal was collected with 4 PMT detectors with the following wavelength emission windows: 425–475 nm for the 405 nm laser, 500–550 nm for the 488 nm laser, 570–620 nm for the 561 nm laser, and 675–725 nm for the 640 nm laser. The specimens were visualised using a 29.9 µm (1.2 airy units) confocal pinhole and a number of z-stacks (typically between 10 and 50), of optical thickness between 0.2–2.0 µm each, were acquired. The fluorescence signal from each z-stack was then projected onto a maximum projection image and used to generate a 3D model of the specimen using Nikon NIS-Elements software (www.nis-elements.com) for Figure 13c. The stacks were further explored and visualised using volume rendering; they were loaded using the open source software Fiji (Schindelin et al., 2012), and the channel with maximum contrast converted to a grayscale TIFF. The resulting TIFF stacks were subsequently loaded in the open source software Drishti (Limaye, 2012), and volume rendered by modifying the 2D histogram transfer function. These renders were used as the basis for Figure 13b, d and for supplementary movie 1.

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

NanoSIMS was performed in the Department of Materials, University of Oxford, using a CAMECA NanoSIMS 50. Regions of interest (ROI) were identified under the optical microscope in polished 30 μm thin sections, and then micro-mapped using bright-field and reflected light. The reflected light images were subsequently used to locate the surface expressions of laminated features within the NanoSIMS. Discs of c. 10 mm diameter containing the ROI were extracted from the thin sections, mounted on NanoSIMS stubs, and coated with a thin (5-10 nm) layer of platinum to provide conductivity at high voltage. Details of qualitative elemental mapping using NanoSIMS in multi-collector mode are given in Wacey et al. (2008) and Kilburn and Wacey (2011). Briefly, a focused primary Cs+ ion beam, with a beam current of 2–4 pA, was rastered over the sample surface, and the sputtered ions were extracted to a double focusing mass spectrometer. Images with sub-100 nm spatial resolution mapping relative ion intensity were acquired over fields of view ranging from 10 μm to 25 μm. Prior to each analysis, the sample area was pre-sputtered to remove surface contamination, implant Cs^+ ions into the sample matrix and attain an approximate steady state of secondary ion emission (cf. Gnaser, 2003). Ion maps of carbon ($^{12}C^-$), nitrogen ($^{12}C^{14}N^-$), silicon ($^{28}Si^-$), sulfur ($^{32}S^-$) and phosphate ($^{31}P^{16}O_2^-$) were then produced simultaneously from the same sputtered volumes of sample. Only relative concentrations of elements can be obtained using this NanoSIMS methodology. Without multiple standards, no inferences can be made from these data concerning either the absolute concentration of elements, or the percentage concentration of one element compared to another.

2.4. Laser Raman Microspectroscopy

Raman was performed in the Centre for Microscopy, Characterisation and Analysis (CMCA), The University of Western Australia, using a *WITec alpha 300RA+* instrument with a *Toptica Photonics Xtra II* 785 nm laser source. Laser excitation intensity at the sample surface was in the 1-5 mW range, well below the intensity that may damage carbonaceous material (e.g., Everall et al., 1991) and comparable to previous studies of the Apex chert (e.g., Olcott Marshall et al., 2012; Sforna et al., 2014). The laser was focused through either a 20x/0.4 or 100x/0.9 objective, with the latter giving a spot size of smaller than 1 μm. Spectral acquisitions were obtained with 600 l/mm grating and a peltier-cooled (-60 °C) 1024 x 128 pixel CCD detector. Laser centering and spectral calibration were performed daily on a silicon chip with characteristic Si Raman band of 520.4 cm^{-1}. Count rates were optimised prior to point spectra acquisition or hyperspectral mapping using the dominant quartz Raman band of 465 cm^{-1}. Spectra were collected in the 100-1800 rel. cm^{-1} region in order that both 1st order mineral vibration modes and 1st order carbonaceous vibration modes could be examined simultaneously. Raman maps were acquired with the spectral centre of the detector adjusted to 944 cm^{-1}, with a motorised stage allowing XYZ displacement with precision of better than 1 μm. Spectral decomposition and subsequent image processing were performed using WITec Project FOUR software, with baseline subtraction using a 3rd or 4th order polynomial. Carbon maps were created by integrating over the ~1600 cm^{-1} 'G' Raman band and quartz maps created by integrating over the 465 cm^{-1} quartz Raman band. The ~1350 cm^{-1} carbon 'D' Raman band was not used to construct maps because this may suffer from interference from the ~1320 cm^{-1} hematite Raman band (cf. Marshall and Olcott Marshall, 2013). Point spectra were acquired using the 100x/0.9 objective, an integration time of 0.5 s and 10 accumulations. All analyses were conducted on material embedded below the surface of the thin section to avoid artefacts in the Raman spectra resulting from polishing and/or surface contamination.

2.5. Energy dispersive elemental mapping (EDS)

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

3. Context

3.1. Regional setting

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

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

3.2. Chinaman Creek Geology

In the vicinity of Chinaman Creek, approximately 5 km west of Marble Bar, thick extrusive accumulations of pillow basalt and komatiite are punctuated by a weathering-resistant ridge of stratiform chert and associated volcaniclastic rocks, informally referred to as the ‘Apex chert’ (Fig. 2). The stratiform chert (unit 4 of Brasier et al., 2005, 2011), the lowermost and thickest of several stratiform cherts in the Apex Basalt (Kato and Nakamura, 2003), is a 10-15 m thick unit of banded iron-rich and iron-poor chert (cf. banded iron formation) of variable texture, composition and colour. Brasier et al. (2005, 2011) recognized that the stratiform chert ridge was separated into three structural blocks (naming them the North, Central and South blocks) by listric normal growth faults. They also showed that, stratigraphically below the ridge, a series of hydrothermal black chert veins cut up through the lower portions of the Apex basalt for up to 1600 m (Figs. 2-3). The veins are particularly thick (up to c. 5 m in diameter) along the growth faults and one such vein (N1 of Brasier et al., 2005; Fig. 3) contains the filamentous microfossil-like artifacts described by Schopf (1993), at a depth of c. 100 m below the palaeosurface. The black chert veins inter-finger with, but do not pass through, the upper stratigraphic limit of the stratiform chert (Brasier et al., 2005; Van Kranendonk, 2006) indicating that they are syndepositional with, or penecontemporaneous to, this unit (Fig. 3). Large clasts of both hydrothermal black chert and stratiform chert are found in the overlying pyroclastic breccia bed (unit 5 of Brasier et al., 2005, 2011) indicating that both were lithified prior to the commencement of the next volcanic cycle. In the vicinity of these veins, the stratiform chert can be highly brecciated with dilatational black chert artificially thickening the stratiform succession and creating angular blocks of bedded chert that appear to ‘float’ in black chert (Brasier et al., 2005), confirming that at least some of the stratiform chert must predate the black chert. Stratiform material may also be found entrained at depth (up to c. 150 m) within the black chert veins, likely caused by either vigorous downward convection during hydrothermal fluid flow or phreatomagmatic explosions (Van Kranendonk, 2006). In contrast to most previous studies that have sought to address the origin of carbonaceous structures in and around the hydrothermal veins (Wacey et al., 2015; Bower et al., 2016 and references therein), here we focus on stratiform chert that shows evidence for sedimentary structures, and mostly crops out some distance away from the major hydrothermal veins (Fig 3).

4. Results

4.1. Petrographic division of stratiform chert types

The stratiform chert is present in each of the three structural blocks at Chinaman Creek, but is most continuous in the north and south blocks. The central block has a comparative paucity of both stratiform and hydrothermal chert. We here divide the stratiform chert at Chinaman Creek into five distinct types on the basis of petrographic observations: silicified volcaniclastics (mostly layered ash and agglomerate; Fig. 4a-b), clotted carbonaceous chert (Fig. 4c-d; cf. Lowe and Knauth (1977), banded microgranular chert (Fig. 4e-f; Supp. Figs. 1-2), metalliferous, mostly iron-rich chert (Fig. 4g-h) and banded black, grey and white chert (Fig. 4i-j).

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

4.2. Microgranular chert fabrics

Fabrics within the microgranular cherts are variable, even on the scale of a single standard thin section and include (i) microgranular zones, (ii) laminated textures and (iii) spherulitic textures, which we address in order below. Additionally, µm-mm scale post-depositional micro-quartz and macro-quartz veins represents multiple later episodes of veining. Brecciation of cherts is common around the large hydrothermal intrusive black chert veins, however, data herein come from non-brecciated stratiform chert away from macro-scale veins.

4.2.1. Microgranular zones and grain types

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

The most common components of the microgranular chert are sub-rounded, carbonaceous lobate grains or ‘fluffs’ (Fig. 5a-b). These range from < 100 µm to > 1 mm in size, with similar sizes of fluffs tending to be spatially correlated, defining discrete domains. The domains are sometimes lense-like indicating a probable sedimentary origin; this might suggest some periodicity to sediment input. Though some fluffs have a high aspect ratio and tapering edges, most are sub-spherical. In all sections, regardless of orientation relative to bedding, these grains have generally cloud-like, ‘fluffy’ morphologies. They frequently have inclusions of wisps of silica and isolated, euhedral-subhedral opaque crystals, which together constitute < 20 % of the grain (Fig. 5a-b). We interpret these fluffy grains as carbon-impregnated silicified volcanic ash (cf. Lowe, 1999; Brasier et al., 2006). Their considerable interparticle (~30%) and intraparticle porosities supports this hypothesis, as does their multiclastic composite constructions (Fig. 5b), suggesting either in-air clotting of multiple ash grains when moistened or submarine or water-surface moistening and coagulation. Elemental mapping shows elevated concentrations of aluminium and potassium within the fluffy grains (Supp. Fig. 3a), also consistent with an origin as volcanic ash (Nakagawa and Ohba, 2003). Furthermore, these grains strongly resemble silicified volcaniclastics described from other members of both the Warrawoona Group and the time-equivalent Onverwacht Group of South Africa (Lowe and Knauth, 1977, 1978; Walsh, 2004; Walsh and Lowe, 1999). Raman micro-spectroscopy confirms the carbonaceous composition of these fluffy grains (Fig. 5c) and shows that carbon impregnation occurred prior to the maximum metamorphic or hydrothermal heating of these rocks in the early-mid Archean (Fig. 5d).

The second most common grain type, accounting for approximately 30% of most microgranular cherts, is the ‘flake’, which is a considerably darker, tapered, elongate grain (Fig. 6a-c). Most flakes are shallowly curved (Fig. 6c); since no way-up criteria are available in what we here interpret as a reworked sediment, no inference of concavity or convexity is implied. Flakes are largely restricted to the microgranular chert, suggesting either a transient formational mechanism, or small reservoir of material from which flaky grains can be drawn, preventing more widespread preservation. Flaky grains appear more densely carbonaceous than fluffy lobate grains, and resemble either: i) ripped-up slivers and laminae of fine-grained sediment (cf. Schieber et al., 2012); ii) ripped-up chips of microbial mats (Noffke, 2010 and references therein); or iii) compressed fluffy grains. Where thin sections are cut perpendicular to the macroscopic banding, there is an obvious preferred orientation of elongate clasts, which is lost in thin sections cut parallel to macroscopic banding. These oriented flakes usually dominate granular layers alternately to the aforementioned fluffs (Fig. 4e). Raman micro-spectroscopy confirms the dominantly carbonaceous composition of flaky grains (Fig. 6a-b), and the carbon ‘D’ and ‘G’ peak intensities, position and shapes of these grains are identical to the aforementioned fluffy grains.

One explanation for the flaky grains is that they are compressed lobate fluffy grains. Rare, partially compressed fluffy grains are observed in silicified ashes at Chinaman Creek; these have tapering margins and jagged bifurcations and compaction is suggested by sutured contacts between adjacent grains (Fig. 6d). However, such compressed lobate clasts with sutured margins have not been observed in the microgranular chert. In addition, when the intra-grain features of known compressed lobate clasts (Walsh and Lowe, 1999) are compared to the flaky grains in the microgranular cherts, the two bear only superficial morphological resemblance. For example, inclusions are rare to absent in the flaky grains described herein, but are common in known compressed lobate clasts. Grain morphologies of the flaky grains suggest a more linear, fissile breakage than the irregular outlines of high aspect ratio lobate grains (Walsh and Lowe, 1999). Flaky grains also appear to lack the enrichment in aluminium and potassium observed in the fluffy grains (Supp. Fig. 3). Furthermore, the occasional occurrence of flaky grains immediately adjacent to uncompressed fluffy grains within a sediment layer (Fig. 6b) discounts a mechanism whereby changes in silicification style may allow one layer to remain uncompressed (fluffy grains) while the next layer becomes compressed (flaky grains).

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

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

4.2.2. Laminated textures

Lamination occurs on a variety of scales. The microgranular cherts are laminated on the mm-scale and these laminae appear to be defined by the relative proportions of fluffy and flaky grains (Fig. 4e, Supp. Fig. 1). Laminae also occur within single, albeit rare, grains in the microgranular chert (Figs. 7-8, Supp. Fig. 2), mostly being defined by the relative proportions of carbon and silica at the tens of microns scale (see section 4.2.2.1 below). Finally, carbonaceous laminae can persist across entire thin sections cut perpendicular to bedding, and can be stacked together vertically for several millimetres in chert zones adjacent to microgranular zones (Fig. 9; see section 4.2.2.2 below). Some of these laminae appear filament-like (cf. Noffke, 2009, 2010) rather than being layered planar surfaces.

4.2.2.1. Laminated grains

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

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

The three-dimensional morphology of carbonaceous material in laminated grains is highlighted by autofluorescence under confocal laser scanning microscopy (CLSM), which demonstrates a wrinkled planar form (Fig. 13; supplementary movie 1) i.e., it is neither filamentous nor tubular. Many of the thicker carbonaceous bands appear multi-laminar in both light microscopy (Fig. 7d) and CLSM (Fig. 13). The largest laminated grain examined by CLSM (from locality CC8 of Brasier et al., 2011) reveals the rolling up of the tapering ends of several laminae; in some cases, these almost roll over by 180˚ (Fig. 13c-d; supplementary movie 1). Rollups such as these have been proposed as evidence for an initial plasticity of structure and are commonly associated with ancient microbial mats (Tice & Lowe, 2004; Tice et al., 2011). Roll-up formation is thought to begin with the erosion, by waves, tides, or other marine current movements, of the original mat. This releases fragments of the mat and leaves tear-up structures in the remaining microbial edifice (Westall et al., 2015). Current movements have a range of erosional outcomes, depending foremost on the morphology and geometry of the microbial mat (Tice et al., 2011). However, if an eroded mat fragment is glutinously bound by cohesive extra-cellular polymeric substance (EPS), current action will deform, but not structurally disintegrate, the fragment, reworking it into a roll-up (cf. Tice and Lowe, 2004).

Secondary laminated grains: Some laminated grains appear secondary in nature and more closely resemble fragmented flakes. Here, micro-quartz veins traverse and intrude the clast disconformably, resulting in sharp contrasts between laminae (Fig. 8a-c). The repetitively alternating banding characteristic of primary fabrics is not present. If these laminae were syn-sedimentary or biologically mediated, a gradation between carbonaceous and siliceous layers would be expected, as observed in the previously described primary grains, and indicative of incremental growth of the precursor sediment and/or biological system. The lamina boundaries here, however, are distinct, signifying brittle breakage or intrusion. Where ‘gradation’ at the margins of laminae is noted, higher magnification observation reveals this to be a result of secondarily loosened darker grains from the adjacent carbonaceous material, likely through pervasive silicification enhancing fracture defects (Fig. 8c). The formational process for these laminated grains is best explained as the intrusion of siliceous fluids into planes of weakness or fracture in the precursor clast, forcing material apart. This process is supported by two further lines of evidence, in addition to the sharp laminar edges. Firstly, the lighter, siliceous bands do not always traverse the entirety of the clast (i.e., there are not always continuous planes as would be expected for a microbial mat or regularly repeating sediment deposition) and are aligned randomly (i.e., they are not likely related to the primary laminated fabric of the microclastic chert). Secondly, the undulations of the carbonaceous laminae appear to be an intimately related response to forcing caused by intercalated growth of silica: there is a near perfect fit between now-separated flakes which leads to our interpretation of these fragments as having once been parts of a larger precursor grain (see particularly the central and lower major grains in Fig. 8a).

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

4.2.2.2. Mesoscopic 'filament-like' laminations

At two localities (CC164 and CCT23 of Brasier et al., 2011), mesoscopic laminations occur in microcrystalline black-grey-white chert zones (we interpret this lithology as an end-member classification of microgranular chert, and it should not be confused with the black-grey-white banded cherts, which are independent) adjacent to microgranular zones. These comprise narrow filament-like textures with an undulating, wrinkled topography (Fig. 9b, dashed arrows). The laminae are stacked vertically into packages varying from < 100 μm to several millimetres thick. Individual filament-like laminae within each package are rather diffuse, but the best-preserved examples are about 10-20 μm thick (Fig. 9d, dashed arrows). The lateral extent of these laminae can be as little as a few tens of micrometers or they may persist across an entire thin section (Fig. 9a) or hand specimen. Laminae are interspersed with microcrystalline silica and this overall texture is sometimes cross-cut by later micro-quartz veins. A number of aggregations of these laminae appear to entrain orientated detrital sediment grains (Figs. 9c-e, 14). Raman micro-spectroscopy confirms that the laminations are carbonaceous, that the carbon is not a modern contaminant, and that the trapped grains are quartz (Fig. 14). This is consistent with the trapping and binding of sediment grains by filamentous microbes and their associated exopolymeric substances, as observed in modern microbial mats, and several fossil examples of Archean age (e.g., Noffke et al., 2001, 2003, 2013; Westall et al., 2011).

4.2.3. Spherulitic textures

The stratiform cherts are often punctuated by spherulitic features. For example, irregular shapes exhibited by some carbonaceous fluffy grains in clotted and microgranular cherts - shapes in marked contrast to the dominant sub-rounded, cloud-like morphologies of these grains - could result from their fragmentation by silica spherules. Fragmentary, homogeneous, and indistinct carbonaceous clasts are often present around lobate grain margins in zones of spherulitic alteration; these may signify the breakup products of larger lobate parent bodies by spherulitic silica growth. These spherulitic textures are often associated with elongate, sometimes partially-filamentous fragments of carbon, that resemble abiogenic pseudofossils previously identified in the black chert veins below the stratiform chert (Brasier et al. 2005, 2011).

5. Discussion

5.1. Assessment of Biogenicity

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

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

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

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

None of the microstructures of interest are found in such late-stage or metastable mineral phases; they occur either in clasts of micro-quartz that were lithified, eroded and reworked prior to the final lithification of the stratiform chert (e.g., Fig. 5a), or are found in primary laminated chert (e.g., Fig. 9). In contrast, spherulitic textures forming filamentous and other carbonaceous fragments are found in void filling cements and cross-cutting veins. These are demonstrably crystal-edge effects, usually resulting from the growth of silica spherules. The carbon in such later phases has been redistributed to such an extent that its morphology cannot be used to determine its origin (cf. Pinti et al., 2009; Wacey et al., 2015).

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

This criterion is met by the sedimentary sequences in the Marble Bar greenstone belt, which have not experienced more than lower greenschist facies regional metamorphism (Hickman and Lipple, 1978; Van Kranendonk, 2006). More specifically, a recent Raman study at the Chinaman Creek locality estimated that the maximum temperatures experienced by these rocks were between 265°C and 360°C, which may represent the peak temperature of regional metamorphism and of hydrothermal fluids respectively (Sforna et al., 2014). Multiple potential biosignatures have been reported previously from similarly silicified sediments of the Pilbara craton (e.g., Sugitani et al., 2010, Wacey et al., 2011; Noffke et al., 2013) indicating that MISS could also be preserved within the stratiform Apex chert.

iv) The geological context of the lithology should be plausible for life; ideally, the lithology should indicate a transgressive depositional phase, since this is the environment in which modern MISS develop.

The stratiform Apex chert is likely to represent a quiescent marine environment in which low-density particles can settle out of suspension, i.e. lobate ash clasts in layers and very fine clay or fragments of microbial mat. Water temperatures would be within the range in which (hyper)thermophilic life could flourish; geochemical evidence, including relatively high concentrations of barium in the stratiform chert and small positive europium anomalies, indicate low-temperature (100-150°C) hydrothermal venting (Kato and Nakamura, 2003). In addition, the input of volcanic detritus may have provided essential elements for life, thus the environment appears habitable. However, further high-resolution sedimentary logging is required to determine whether a transgression can be identified within the stratiform chert unit. This work should also search for evidence of macroscopic MISS that are as yet unreported from this unit.

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

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

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

Microbial mats are demonstrably one of the most ancient and enduring biological communities, with evidence of microbial mat/sediment interaction reported from rocks of similar age to the Apex chert in both the Pilbara and Barberton regions (e.g., Tice and Lowe, 2004; Allwood et al., 2006; Westall et al., 2001, 2006, 2015; Noffke et al., 2013). An interpretation of the laminated and filamentous Apex microstructures as MISS fits within this evolutionary context and, by analogy to modern ecosystems, implies community behavior of a microbial mat (Noffke, 2008; Schieber et al., 2007). We do not interpret any of the microstructures as microfossils, though we raise the possibility that the filament-like laminae entraining sediment grains may be the diffuse remnants of mat-building filamentous microorganisms (cf. Noffke et al., 2003). Further detailed geochemical and morphological research must be conducted into these, however, before such an interpretation can be substantiated.

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

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

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

In addition to the wavy and wrinkled laminae described above, some of the putative MISS contain grains around which the filament-like laminae wrap. In modern biofilms, microbial mats and stromatolites, such microstructures form via the trapping and binding of sediment grains by 'sticky' filamentous microorganisms and their associated extra-cellular polymeric substances (EPS; e.g., Reid et al., 2000). Similarly, rolled-up microstructures in modern settings are cited as evidence of an initial plasticity, hence cohesiveness in the sediment, and ancient examples have been interpreted as resulting from the interaction of erosive forces, such as currents, with a microbial mat (e.g., Tice and Lowe, 2004). We have identified putative roll-ups in the microclastic cherts (CC8 and CCT27), though they are of smaller size than well-accepted Archean examples (Tice and Lowe, 2004). Additionally, a plausible interpretation of the flake-like grains reported here is that they are micro-scale analogues of microbial mat chips commonly found in modern MISS assemblages, also formed when water agitation (driven by tides or storms, for example) tears small pockets of semi-cohesive material from their parent microbial mat (Gerdes and Krumbein, 1987, Tice et al., 2011).

x) MISS should possess geochemical signals indicative of biology

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

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

5.2. Summary of potential biogenicity

Of the range of textures exhibited by the stratiform cherts, we find that it is the microgranular cherts that hold the most promise for the retention of biosignatures. Primary laminated grains, filament-like wrinkle structures, roll-ups, and flaky grains may all plausibly be interpreted as remnants of a microbial mat community. The texture of the microclastic cherts suggests a shallow, quiescent environment of deposition, in which weak but persistent currents facilitated the orientation and imbrication of elongate clasts. There is a prominent volcaniclastic component to the microgranular cherts, and evidence of a proximal silicic source comes in the form of interbedded silicified ashes and other volcaniclastics that constitute much of the stratiform stratigraphy at Chinaman Creek (Kato and Nakamura, 2003; Brasier et al., 2011). It may be that this volcanic input provided elements, particularly metals, significant to the emergence of life in this habitat (cf. Barras, 2012; Van Kranendonk, 2006). However, the lack of evidence for macroscopic MISS or definitive microfossils in this unit urges a note of caution. In addition, the environment of deposition and hydrothermal style of silicification (e.g., Kato and Nakamura, 2003), overprinting some of the primary sedimentary features, is rather different from that of traditional siliciclastic settings in which MISS are well understood (cf. Noffke, 2010), and for which the criteria outlined above were primarily designed.

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

This study adds to the growing evidence for a diversity of primitive life in the early Archaean era, and provides a detailed assessment of carbonaceous microstructures within the stratiform Apex chert at Chinaman Creek, a lithology that has been only very briefly described in previous work (e.g., Brasier et al., 2011; Sforna et al., 2014). Our data support the hypothesis that shallow-water environments, together with input from volcanic and hydrothermal sources, were likely pivotal niches occupied by simple, prokaryotic mat-forming organisms. The MISS-type structures described herein are found in relatively close proximity to penecontemporaneous hydrothermal fabrics, yet have no apparent genetic association with these higher-temperature events; thus their interpretation as biological signals is more probable than an abiotic origin.

5. Conclusions

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

The macroscopic sedimentary textures of the stratiform Apex cherts, which have dominant silt-grade grains in most localities studied, and occasional imbrication of semi-lithified chert fragments, hints at a shallow marine depositional environment. The poor lateral continuity of individual chert layers argues against deep marine pelagic settling of sediment. The depositional environment is herein interpreted to be a protected shallow marine environment, where weak currents are intermittently present to sort and orient clasts. Inter-bedded metalliferous cherts, either ferruginous or jaspilitic, may signify a change in redox state or temperature, most likely linked to the penecontemporaneous hydrothermal venting and the intrusive black chert veins (Kato and Nakamura, 2003; Brasier et al., 2005, 2011). Volcanic components, largely ashfall, are prevalent throughout the stratiform sequence, consistent with a shallow-marine environment situated adjacent to active volcanoes. This is similar to the environmental setting described for other units from both the Pilbara and Barberton regions containing putative biological remains of approximately equivalent age (e.g., Kitty's Gap Chert and Josefsdal Chert; Westall et al., 2006, 2011, 2015). The stratiform Apex chert depositional environment is not compatible with the deeper marine regime suggested for some Archean cherts (Lowe, 1984; Paris et al. 1985).

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

CLSM is here shown to be an effective technique for the imaging of carbonaceous microstructures in these Archean cherts; the autofluorescence of carbonaceous material produces sequential tomograms through the depth of all thin sections investigated herein. This technique enables the generation of sub-micrometer-scale spatial resolution, three-dimensional renderings of features of biogenic interest, and here strengthens the morphological evidence for the biogenicity of some of the described laminated carbonaceous textures. NanoSIMS and laser Raman provide evidence for the concentration of life-significant elements in microstructures that closely resemble the morphology of microbialites. An encouraging combination of both morphology and chemistry pertinent to life in the laminated microstructures of the stratiform cherts, a lithology representing a geologically plausible environment for microbial life, suggests that such features have a biological origin. Although no compelling evidence for life has been found in the underlying heavily studied black chert veins (Brasier et al., 2002, 2005, 2006, 2011, 2015; Wacey et al., 2015; Bower et al., 2016), evidence of an early Archean biological community may yet be present at Chinaman Creek.

Acknowledgements

We are grateful for the assistance of Owen Green and Jeremy Hyde at Oxford University Department of Earth Sciences for the preparation of thin sections for microscopy and discs for NanoSIMS. KHL was supported by Oxford University Department of Earth Sciences master’s thesis funding, St Edmund Hall, and by a Gareth Roberts grant. We acknowledge the facilities, scientific and technical assistance of the Australian Microscopy & Microanalysis Research Facility at the Centre for Microscopy Characterisation and Analysis, The University of Western Australia, a facility funded by the University, State and Commonwealth Governments. DW was funded by the European Commission and the Australian Research Council (FT140100321). This is Australian Research Council Centre of

Excellence for Core to Crust Fluid Systems publication number XXX (to be filled in on acceptance). Chris Grovenor at Oxford University Department of Materials kindly provided advice and consultation with regard to NanoSIMS and arranged our access to the facilities of the Department of Materials at Begbroke Park, Oxford. RG is a Scientific Associate at the Natural History Museum, London, and a member of the Interdisciplinary Centre for Ancient Life (UMRI), and was an 1851 Royal Commission Research Fellow for the majority of this project.

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Figure Captions

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

Figure S1. Textural characteristics of microclastic cherts at Chinaman Creek. a) Microclastic chert (sample CC8) is unequivocally layered: in this scan of part of a thin section, alternating darker and lighter coloured bands can be observed, which indicate layers with a predominance of lobate fluffy ash clasts and elongate 'rip-up' flaky clasts, respectively. b) A second microclastic chert (sample CCT27) also consists of darker ash grains and lighter layers dominated by silica and flaky clasts, though here the banding is less distinct. There is a general coarsening-upward trend in grain size here. c) Microcrystalline and megaquartz veins regularly cut across thin sections but are rarely parallel to bedding. d) General texture of the microcrystalline cherts, showing a relatively well-sorted, clast-supported texture. Scale bar is 5 mm in (a) and (b) and 500 µm in (c) and (d).

Figure S2. Variations of microclastic chert at Chinaman Creek. a) Slide scan of microclastic chert (sample CC43), which is rich in orientated pale clasts, some of which are banded. (b-c) Examples of banded clasts from sample CC43. Scale bar equals 3 mm in (a) and 500 µm in (b-c).

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

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