



# In search of early life: Carbonate veins in Archean metamorphic rocks as potential hosts of biomarkers



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## ABSTRACT

The detection of early life signatures using hydrocarbon biomarkers in Precambrian rocks struggles with contamination issues, unspecific biomarkers and the lack of suitable sedimentary rocks due to extensive thermal overprints. Importantly, host rocks must not have been exposed to temperatures above 250 °C as at these temperatures biomarkers are destroyed. Here we show that Archean sedimentary rocks from the Jeerinah Formation (2.63 billion yrs) and Carawine Dolomite (2.55 billion yrs) of the Pilbara Craton (Western Australia) drilled by the Agouron Institute in 2012, which previously were suggested to be suitable for biomarker studies, were metamorphosed to the greenschist facies. This is higher than previously reported. Both the mineral assemblages (carbonate, quartz, Fe-chlorite, muscovite, microcline, rutile, and pyrite with absence of illite) and chlorite geothermometry suggest that the rocks were exposed to temperatures higher than 300 °C and probably ~400 °C, consistent with greenschist-facies metamorphism. This facies leads to the destruction of any biomarkers and explains why the extraction of hydrocarbon biomarkers from pristine drill cores has not been successful. However, we show that the rocks are cut by younger formation-specific carbonate veins containing primary oil-bearing fluid inclusions and solid bitumens. Type 1 veins in the Carawine Dolomite consist of dolomite, quartz and solid bitumen, whereas type 2 veins in the Jeerinah Formation consist of calcite. Within the veins fluid inclusion homogenisation temperatures and calcite twinning geothermometry indicate maximum temperatures of ~200 °C for type 1 veins and ~180 °C for type 2 veins. Type 1 veins have typical isotopic values for reprecipitated Archean sea-water carbonates, with  $\delta^{13}\text{C}_{\text{VPDB}}$  ranging from  $-3\text{‰}$  to  $0\text{‰}$  and  $\delta^{18}\text{O}_{\text{VPDB}}$  ranging from  $-13\text{‰}$  to  $-7\text{‰}$ , while type 2 veins have isotopic values that are similar to hydrothermal carbonates, with  $\delta^{13}\text{C}_{\text{VPDB}}$  ranging from  $-18\text{‰}$  to  $-4\text{‰}$  and  $\delta^{18}\text{O}_{\text{VPDB}}$  ranging from  $-18\text{‰}$  to  $-12\text{‰}$ . Evidently, the migration and entrapment of hydrocarbons occurred after peak metamorphism under temperatures congruous with late catagenesis and from fluids of different compositions. The relatively high temperatures of vein formation and the known geotectonic history of the rocks analysed suggest a probable minimum age of 1.8 billion yrs (Paleoproterozoic). Our results demonstrate that post peak-metamorphic veins provide an exciting opportunity in the search for evidence of early life. The integration of petrological and organic geochemical techniques is crucial for any future studies that use biomarkers to reconstruct the early biosphere.

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## 1. Introduction

The search for evidence of life in the Archean eon (>2.5 billion yrs), its composition and impact on Earth's environment relies on the occurrence of microfossils, stromatolites, and on the analysis of stable isotopes and hydrocarbon biomarkers

(Dutkiewicz et al., 2006; Waldbauer et al., 2009; Kamber et al., 2014; Planavsky et al., 2014; Brasier et al., 2015; Stüeken et al., 2015). Biomarkers are organic compounds that have particular biosynthetic origins and are preserved as part of the organic matter of sediments and sedimentary rocks (Killops and Killops, 2009) and can be used as a source of information, especially where microfossils are absent. However, the field of Archean biomarker research has recently encountered major pitfalls. In a recent study to reappraise biomarkers believed to be indigenous

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to Archean sedimentary rocks from the Pilbara Craton (Western Australia; Brocks et al., 1999, 2003; Eigenbrode et al., 2008). French et al. (2015) demonstrated that previously detected steranes and hopanes indicative for eukaryotes and bacteria were, in fact, the result of sample contamination. This had been suggested by an earlier study of the carbon isotopic composition of the kerogen and extracted hydrocarbons (Rasmussen et al., 2008). These biomarkers previously were considered to support the presence of oxygenic photosynthesis prior to the Great Oxidation Event (GOE) ca. 2.45 billion yrs (Ga) ago, and coincided well with inorganic evidence for “whiffs” of oxygen before oxygen became fully retained in the atmosphere (e.g., Anbar et al., 2007; Planavsky et al., 2014). Consequently, these biomarkers can no longer provide evidence supporting the rise of oxygen-producing bacteria (cyanobacteria) and eukaryotes before the GOE. The rock samples investigated by French et al. (2015) contained ample organic material (up to 6.7% total organic carbon; French et al., 2015), but regional metamorphism was the most likely explanation for the absence of any detectable indigenous biomarkers, with only highly thermally stable hydrocarbons remaining. Indeed, hydrocarbon biomarkers can be destroyed when exposed to pressures and temperatures consistent with metagenesis (Hunt, 1996). This severely limits the search for Archean biomarkers, since all Pilbara Craton sedimentary rocks from that time were metamorphosed during two major thermo-tectonic events at 2.430–2.40 Ga and 2.215–2.145 Ga (Rasmussen et al., 2005).

A further complication in the reconstruction of the Archean biosphere and environments is the prevalence of “non-specific biomarkers”. For example, the most prominent hydrocarbon biomarker for cyanobacteria, 2-methylhopanoid (Summons et al., 1999), has been found in other bacteria strains, and therefore it is not exclusive to cyanobacteria nor suitable for investigating oxygenic photosynthesis (e.g., Rashby et al., 2007; Welander et al., 2010). On the positive side, non-exclusive hydrocarbon biomarkers for cyanobacteria include mid-chain methylheptadecanes (Schirmer et al., 2010), and high abundances of these molecules might still suggest oxygenic photosynthesis. Other biomarkers that would indicate the presence of oxygen include any type of alkylated steranes, which are derived only from eukaryotes and require molecular oxygen for their biosynthesis (Volkman, 2005). Whether eukaryotes were already present in the Archean or evolved later is not known, as the previously detected alkylated steranes in the Pilbara rocks have now been shown to reflect contamination (Rasmussen et al., 2008; French et al., 2015). One of the most persistent complications in the detection of early life using biomarkers is the introduction of hydrocarbons by, for example, more recent oil migration, or during sampling and handling of rock samples, thus requiring careful identification of these contaminants (e.g., Rasmussen et al., 2008; Brocks, 2011).

One way to minimise contamination issues is to analyse oil trapped in fluid inclusions. These oil-bearing fluid inclusions are normally hosted within sealed cavities in mineral grains such as calcite, dolomite, feldspar, and quartz, making them relatively stable when exposed to high temperatures and pressures (~350 °C, 2 kbar; e.g., Dutkiewicz et al., 2006; George et al., 2008, 2012). Oil-bearing fluid inclusions have previously been found in a range of Precambrian rocks (Dutkiewicz et al., 1998, 2006; George et al., 2008). They are protected from the degradation processes that can otherwise affect oil in an open pore space, partly because they are closed systems with high fluid pressures, and partly because they contain no clays or other minerals or metals that might catalyse oil-to-gas cracking (George et al., 2008). The included oil thus remains relatively unaltered compared to its host rock, and examples have been successfully analysed in numerous hydrocarbon biomarker studies (e.g., Dutkiewicz et al., 2006; George et al., 2008,

2012). The main problem with the interpretation of oil-bearing fluid inclusion geochemistry is to determine the timing of trapping of hydrocarbon fluids (George et al., 2012).

Nonetheless, the biggest challenge in our view is to find suitable Archean rocks that have experienced adequately low metamorphic grades throughout their geological history such that biomarkers remain intact. Many studies that reported the presence of syngenetic biomarkers in Archean rocks (Brocks et al., 1999, 2003; Eigenbrode et al., 2008; Waldbauer et al., 2009) would have benefited from additional petrological data. As biomarkers can be preserved in different mineral phases or in fractures and cracks (e.g., Nabbefeld et al., 2010; Brocks, 2011) and could therefore represent different origins, it is essential to know the structure of the sample in order to correctly interpret any biomarker data obtained from bulk analysis. With additional petrological data, such as mineral assemblages, an organic geochemist can assess if metamorphosed rocks can still theoretically contain biomarkers before the rocks are analysed for biomarkers using techniques that are relatively expensive and time-consuming. With this supporting information it may be much easier to assess any potential contamination problems and to provide independent constraints on the relative timing of oil generation and migration. For example, one would not expect biomarkers to persist in rocks that have been heated to temperatures in excess of ~250 °C (e.g., Hunt, 1996).

In this study, we document the presence of oil-bearing fluid inclusions and solid bitumens in carbonate veins of new ultra-clean drilled Archean rock samples from the Pilbara Craton. Our approach includes an extensive petrological characterisation of sedimentary host rocks, veins and oil-bearing fluid inclusions that reveal different grades of metamorphism between the host rock and the veins, making the oil-bearing fluid inclusions and solid bitumens promising targets for biomarker analyses.

#### Geological setting

The Agouon Institute Drilling Program (AIDP) drilled three ca. 300 m-long cores in the Pilbara Craton in 2012 (Fig. 1; French et al., 2015), in order to obtain fresh, unadulterated Archean rocks for helping to unravel early life signatures during the Archean using hydrocarbon biomarkers, light stable isotopes, transition metal isotopes, and redox-sensitive detrital minerals.

Core AIDP-1 was drilled in the Coonterunnah Subgroup of the Warrawoona Group, Pilbara Supergroup (21°06′38″S, 119°06′4″E), and includes the metamorphosed volcanic Coucal Formation (3.52 Ga; Van Kranendonk et al., 2007). As this core was drilled as a negative control sample for biomarkers and does not contain oil-bearing fluid inclusions it will not be discussed further (see S-Fig. 1 for a detailed AIDP-1 sample description). Core AIDP-2 was drilled in the Ripon Hills region (21°16′51″S, 120°50′2″E) and core AIDP-3 was drilled in the Tunkawanna region (21°46′32″S, 117°34′11″E; Fig. 1A,B). Core AIDP-2 represents a relatively shallow water facies and includes the Carawine Dolomite (2.55–2.54 Ga) of the Hamersley Group that conformably overlies the Jeerinah Formation (2.69–2.63 Ga) of the Fortescue Group. AIDP-3 is a time-equivalent core in a deeper water facies compared to AIDP-2, and includes the Marra Mamba Iron Formation (~2.60 Ga) of the Hamersley Group, conformably overlying the Jeerinah Formation (French et al., 2015). Both cores were drilled in areas where syngenetic biomarkers of Archean age were reported at the same stratigraphic levels (e.g., Brocks et al., 1999; Eigenbrode et al., 2008), and where the metamorphic facies was perceived to be adequately low grade for biomarkers to be preserved (prehnite-pumpellyite facies: <300 °C, <7 kbar; e.g., Smith et al., 1982; French et al., 2015). The Carawine Dolomite and Jeerinah Formation are the primary targets of this study as they appear to contain

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