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# Archean (3.33 Ga) microbe-sediment systems were diverse and flourished in a hydrothermal context

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## ABSTRACT

**Interacting, diverse microbe-sediment systems exist in natural environments today but have not yet been recognized in the oldest records of life on Earth (older than 3.3 Ga) because of lack of distinctive biomarker molecules and patchy preservation of microbial paleocommunities. In an in-situ outcrop- to microbial-scale study, we have differentiated probable phototrophic, chemolithotrophic, and chemo-organotrophic fossil microbial signatures in a nearshore volcanogenic sedimentary setting in 3.33 Ga rocks of the Josefsdal Chert, Barberton greenstone belt, South Africa, while demonstrating the importance of contemporaneous hydrothermal activity. Hydrothermal fluids, as a nutrient source, strongly controlled the development and distribution of the microbial communities and, as a silicifying agent, contributed to their rapid fossilization. We thus show that intricate microbe-sediment systems are deep-rooted in time and that at least some early life may indeed have been thermophilic.**

## INTRODUCTION

Microbial communities in natural environments exist as multispecies assemblages that interact directly with one another and with their surroundings, and thus can be viewed as distinctive systems (Nealson, 1997). For example, a wide diversity of organotrophic and lithotrophic (chemotrophic) microorganisms has been described in present-day, carbon-rich hydrothermally influenced basaltic sediments (Callac et al., 2013), similar to those described from early Earth. However, lack of specific biomarker preservation in very ancient rocks (Summons, 1993) and haphazard preservation of microbial communities in general (Campbell et al., 2001; Orange et al., 2009) make tracing such systems in fossilized form to the oldest records of life on Earth (early Archean, older than 3.3 Ga) very challenging. Nevertheless, phototrophic microbial communities in early to mid-Archean (ca. 3.5–3.2 Ga) strata of South Africa and Australia have been well documented (Tice, 2009; Allwood et al., 2006; Heubeck, 2009; Westall et al., 2006, 2011a, 2011b). On the other hand, no previous investigations have recognized and addressed the syngenetic diversity of Archean microbial paleocommunities—both phototrophs and che-

motrophs—within their sedimentary habitats at the microbial scale and using in-situ methods. Here we describe a macroscopic to microscopic investigation of the sedimentary and geochemical settings of widespread, fossilized phototrophic and chemotrophic microorganisms in early to mid-Archean (3.33 Ga) coastal sediments from the Josefsdal Chert, Barberton greenstone belt, South Africa. We also emphasize the importance of contemporaneous hydrothermal activity both as a source of energy for biomass production and as the means of preserving the biosignatures.

## MATERIALS AND METHODS

Field investigations to study and sample the Josefsdal Chert (25°57.949'S, 31°04.712'E; Fig. 1) were carried out between 1999 and 2014, with detailed maps and stratigraphic sections constructed during the 2012 and 2014 campaigns. Raman analyses were completed at the Centre de Biophysique Moléculaire–Centre National de la Recherche Scientifique (Orléans, France) on polished thin sections using a WITec Alpha500 RA Raman spectrometer (Foucher and Westall, 2013). Whole-rock analyses were conducted utilizing laser ablation–inductively coupled plasma–mass spectrometry (LA-ICP-MS) and ICP–optical emission spectrometry (ICP-OES) at the University of Cardiff (UK). In-situ elemen-

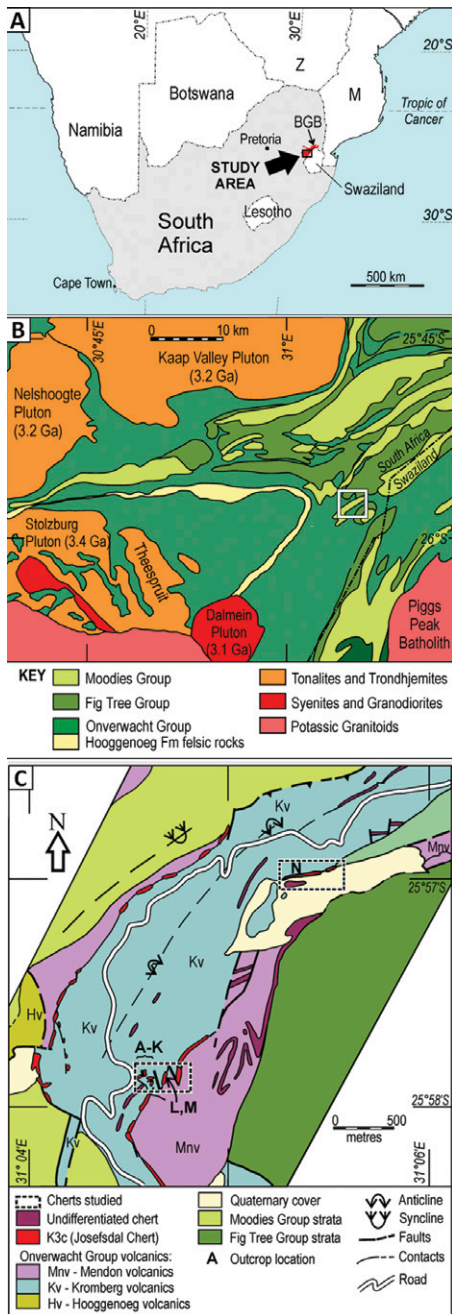
tal mapping was undertaken on 60–80- $\mu$ m-thick rock sections by proton induced X-ray emission (PIXE) spectrometry at the Applications Interdisciplinaires des Faisceaux d'Ions en Région Aquitaine (AIFIRA) facility, Centre des Etudes Nucléaires de Bordeaux-Gradignan (France). In-situ  $\delta^{13}\text{C}$  and  $\delta^{34}\text{S}$  measurements were made on polished rock surfaces and thin sections using a Cameca 1280 HR microprobe at the Centre de Recherches Pétrographiques et Géochimiques (Vandœuvre-lès-Nancy, France). Details of the operating conditions for the various analytical techniques can be found in the Methods section of the GSA Data Repository<sup>1</sup>.

## SHALLOW-WATER SEDIMENTARY CONTEXT

The early Archean Josefsdal Chert comprises a thin package (6–30 m) of silicified, mainly sandy

<sup>1</sup>GSA Data Repository item 2015215, methods; Figure DR1 (correlation of stratigraphy measured in the outcrops shown in Figure 1C); Figure DR2 (comparison of sedimentary structures from the Josefsdal chert [Figs. 2B–2E] with analogous Phanerozoic structures); Figure DR3 (geochemical data [PIXE, Raman] documenting a hydrothermal trace element signature [As, Ni, Cu, Zn] in the siliceous cement of Facies D sediments and associated biofilm); Figure DR4 (soft sediment deformation in a phototrophic biofilm); Figure DR5 (evidence for syngenetic and early diagenetic hydrothermal silica infiltration); Figure DR6 (micrographs and Raman carbon map of Type 1 phototrophic biofilm [cf. Figs. 3A–3D, 3G]); Figure DR7 (comparison of Type 1 films and Type 2 clots [cf. Fig. 3] from Josefsdal with Jurassic hydrothermal extreme environment analogs); Figure DR8 (sketch of the inferred environment of deposition of the different facies [A–D] [Fig. 2]); Table DR1 (Josefsdal Chert sedimentary facies and hydrothermal and microbial associations); and Table DR2 (geochemical data from the Josefsdal Chert samples), is available online at [www.geosociety.org/pubs/ft2015.htm](http://www.geosociety.org/pubs/ft2015.htm), or on request from [editing@geosociety.org](mailto:editing@geosociety.org) or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

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**Figure 1. Location and geological context of Josefsdal Chert, Barberton greenstone belt, South Africa. A:** Study area, Barberton greenstone belt (BGB, red). Z—Zimbabwe; M—Mozambique. **B:** Geological map of BGB (after de Ronde and de Wit, 1994). Study area shown by white box. Fm—Formation. **C:** Detailed geological setting of Josefsdal Chert (K3c) (Lowe et al., 2012); boxes indicate studied outcrops, A–N. Curved arrow in Moodies Group indicates overturned syncline, and in the kromberg volcanics, an overturned anticline.

to silty volcanogenic sediments sandwiched between thick basaltic volcanic sequences (Lowe, 1999) (Figs. 1 and 2A). Mapped as the 3.33 Ga (Byerly et al., 1996) K3c chert (Lowe et al., 2012), the Josefsdal Chert crops out over a distance of several kilometers (Fig. 1). Four

stratigraphic units (1–4) containing four interbedded sedimentary (chert) facies associations (A–D) were deposited under shifting energy conditions in a nearshore paleoenvironment (Fig. 2). The basal portion (unit 1) comprises swaley to hummocky, cross-stratified, well-sorted sandstone representing upper offshore to shoreface storm deposits (Dumas and Arnott, 2006) (facies A; Fig. 2B). These strata grade upward into red, iron-stained, parallel-laminated, poorly sorted sand and mud in thickening-thinning, rhythmic packages (facies B; Fig. 2C), indicative of a tidally influenced, fair-weather shoreface setting and dominating unit 2. Unit 2 is capped by a distinctive black and white banded chert (facies C; Fig. 2D).

The upper portion of the studied sequence (units 3 and 4) constitutes alternations of parallel-laminated to rippled—some oscillatory—sandstone (facies D; Fig. 2E), swaley to hummocky cross-stratified sandstone (facies A), and facies C, developed in shoreface to foreshore settings. Rare desiccation cracks in facies A point to occasional subaerial exposure. Uniform, parallel laminae and thin beds of graded lapilli, signifying individual volcanic episodes, are particularly common in the green and black cherts of facies D (Fig. 2E) and allow very accurate local correlation to be established between outcrops A–N (Fig. 1C; Fig. DR1 in the Data Repository). See Table DR1 in the Data Repository for a detailed description of the sedimentary facies and Figure DR2 for field photographs with comparisons to analogous Phanerozoic shallow-marine sedimentary structures.

### PENECONTEMPORANEOUS HYDROTHERMAL ACTIVITY

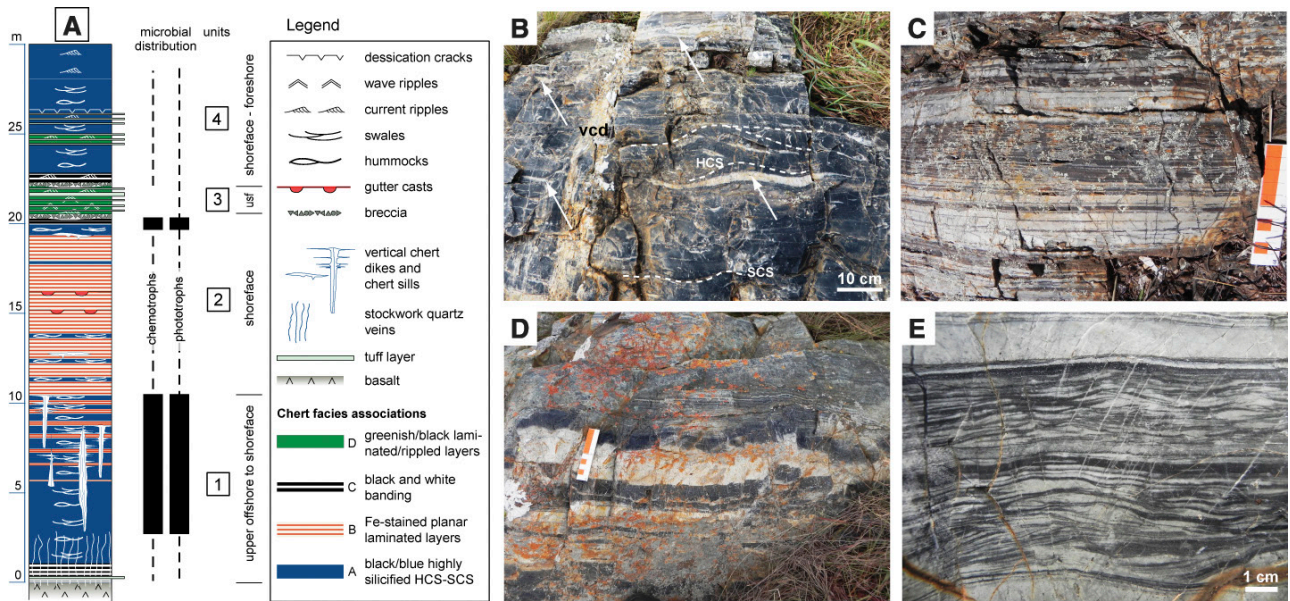
There is significant evidence for paleohydrothermal activity in the Josefsdal area, its timing estimated to have ranged from contemporaneous with sedimentation to early post-lithification. The pervasive silicification of Josefsdal sediments (95% to >99% SiO<sub>2</sub>) and the immediately underlying basalts was due to pore water circulation of hydrothermal fluids enriched in silica originating from Si-enriched seawater–rock interactions, as indicated by their trace element signature (e.g. As, Ni, and Cu; Fig. DR3), Mud of Queensland (MUQ)–normalized (Bolhar et al., 2005) rare earth element + Y patterns, and positive Y and Eu anomalies (Table DR2) (cf. Hofmann and Harris, 2008). Soft deformation of the finely laminated photosynthetic microbial biofilms (Fig. DR4) and of some intercalated horizons of clear silica (Fig. DR5) are evidence for the contemporaneous injection of hydrothermal fluids and/or hydrothermal trace element–enriched pore waters. The lack of compaction and exceptional preservation state of the photosynthetic biosignatures (Fig. 3; Fig. DR4) indicate that polymerization of the siliceous pore waters was rapid. Some thin beds of platy intra-

formational conglomerate developed in situ, and may have been influenced in their formation by hydrothermal fluid injections which, in places, also infiltrated into partially lithified (silicified) sediment layers (e.g., Figs. 2B and 2D; Fig. DR5A), locally forming botryoidal chert sills up to 10 cm thick. Moreover, vertical, white to translucent chert dikes (up to 40 cm wide and up to 10 m long) indicate slightly to somewhat later fluid injection into lithified, lower (facies A only) Josefsdal sediments (Fig. 2B). At Josefsdal, our results are consistent with evidence for Archean hydrothermal activity driven by the circulation of Si-enriched seawater through cooling lavas as seen elsewhere in the Barberton greenstone belt (Hofmann and Harris, 2008) and the Pilbara (Australia; e.g., van den Boorn et al., 2007).

### IN-SITU MICROBIAL COMMUNITIES AND DETRITAL CARBON

In the Josefsdal hydrothermal, shallow-marine depositional setting, we document three types of occurrences of carbonaceous material using a multiscale, in-situ approach (Fig. 3). Type 1 consists of layered packets (100–1000 μm thick) of laterally extensive (several centimeters), wavy, thin carbonaceous films (~10 μm to a few tens of micrometers) that commonly incorporate trapped particles of carbonaceous or mineral detritus (Figs. 3A–3C) (Westall et al., 2006, 2011a). The films occur at the tops of sandy hummocks in facies A and on bedding planes in facies B–D, evidently formed on the seabed during quiet intervals between storms. The films also occur in clear chert horizons inferred as silica gel of hydrothermal origin (facies A, exposures M–L, and facies C, exposure H) (see Figs. DR4 for additional images of the type 1 films, Figs. DR6A–DR6D for Raman carbon-mineral maps, and Figs. DR7B and DR7F for comparison to Jurassic hydrothermal analogs). The occurrence of carbonaceous films in the photic zone, their morphological characteristics, and in-situ-measured stable carbon isotope ratios of –22‰ to –13‰ δ<sup>13</sup>C<sub>VPDB</sub> (Vienna Peedee belemnite) are consistent with the type 1 films representing microbial mats utilizing a phototrophic metabolism (cf. Tice, 2009). The previously reported association of one of these biofilms with chemorganotrophic sulfate-reducing bacteria (SRB) (Westall et al., 2011a), the activities of which induced still-preserved in-situ calcification, is supported in this study by in-situ measurements on the same fossil biofilm yielding δ<sup>13</sup>C<sub>VPDB</sub> signatures as light as –45‰ and depleted δ<sup>34</sup>S<sub>CDT</sub> (Canyon Diablo troilite) values of –24‰ (cf. Thomazo et al., 2013). Microprobe mapping of Ca associated with other Josefsdal type 1 biofilms (data not shown) suggests that the association of SRBs with phototrophs is widespread.

Type 2 carbonaceous occurrences are clots (cf. Bailey et al., 2009) formed as carbon coatings on sand-sized (50–500 μm) volcanic clasts



**Figure 2. Stratigraphy and facies associations of Josefsdal Chert, South Africa.** Stratigraphic column (A; based on outcrops L–M) comprises four stratigraphic units (1–4) consisting of four interbedded sedimentary (chert) facies associations (A–D) that represent sedimentation in shifting upper offshore to foreshore environments. B: Swaley and hummocky cross-stratification (SCS-HCS) (facies A), deposited by storms in the upper offshore to shoreface, dominate Unit 1, which also exhibits abundant, penecontemporaneous, hydrothermal white to translucent, vertical chert dikes (vcd) and thin chert sills. C: Iron-stained, poorly sorted, rhythmically laminated sediments of facies B, interpreted to have been deposited in a shoreface setting that was periodically tidally influenced. D: Hydrothermal black and white banded chert constitutes facies C, which caps unit 2. E: Planar-laminated volcanic accretionary lapilli and ash (facies D; cf. Lowe, 1999), comprising Unit 3, were intermittently reworked into small current and wave ripples. Facies D is inferred to have been deposited in an upper shoreface (usf in A) setting. Unit 4 contains alternations of facies A (with rare dessication cracks), C, and D that accumulated in shoreface to foreshore settings. Biosignatures occur ubiquitously in all facies; they are generally uncommon but dense in the vicinity of paleo-hydrothermal activity.

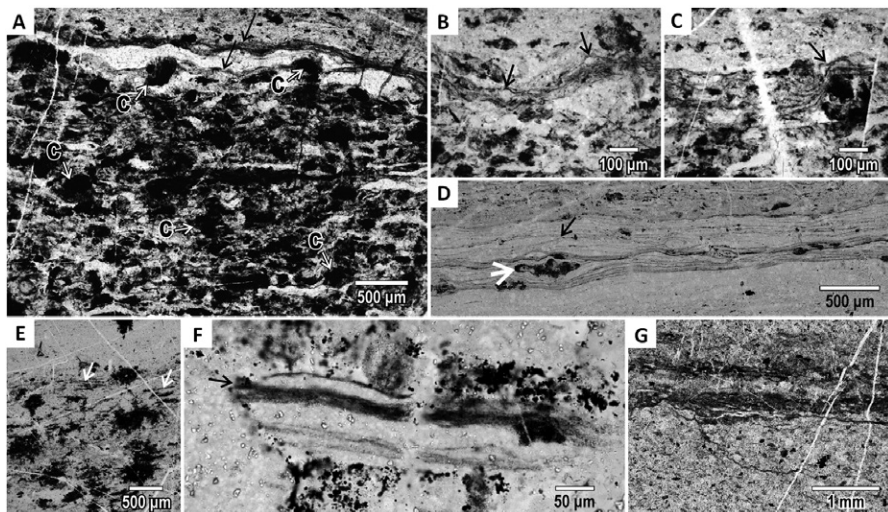
(Fig. 3A; Figs. DR7C and DR7D) or irregular (cauliflower-shaped or spiked), 200–500 μm features (Fig. 3E) that appear to float within an inferred hydrothermal silica gel. In the vicinity of hydrothermal effusions (e.g., outcrops H and K–M), they are densely packed (Fig. 3A) and co-occur with the phototrophic type 1 films (Figs. DR6 and DR7A; and Fig. DR7B for comparison to Jurassic hydrothermal analogs). Type 2 clots apparently formed in situ as indicated by a lack of evidence for transport, their delicate morphologies, and spatial association of many examples

with the surfaces of detrital volcanic grains (cf. Westall et al., 2011b), the latter similar to microbial colonization of younger hydrothermally influenced deposits (Campbell et al., 2001; Callac et al., 2013). High concentrations of Josefsdal clots at sites of greatest hydrothermal activity (Fig. 3A; Fig. DR7A) suggest thermal fluids as a principal energy source, providing inorganic (e.g., H<sub>2</sub>, CO<sub>2</sub>) or organic compounds (e.g. fatty acids, alcohols, ketones) to fuel chemolitho- and chemo-organotrophic metabolisms (cf. Sleep and Bird, 2007; Callac et al., 2013).

Type 3 carbonaceous material occurs as transported detrital particles, such as fragments of finely laminated type 1 films (e.g., Fig. 3F), as elongated flocs of type 2 clots (white arrow of Fig. 3D), or very fine-grained detritus (e.g., black arrow of Fig. 3D).

**DIVERSE ARCHEAN MICROBE-SEDIMENT SYSTEM, BIOSIGNATURE PRESERVATION, AND THE IMPORTANCE OF HYDROTHERMAL ACTIVITY**

The distribution and preservation of fossil microbial phototrophic biofilms and chemot-



**Figure 3. Types of carbonaceous material in the Josefsdal Chert, South Africa.** A: Dark wavy, phototrophic laminae (type 1; black arrows) coexisting with dark, chemotrophic clots (type 2; labeled as C). B,C: Details of phototrophic biofilms showing entrapped detrital grains (arrows, B) and compensation of laminae over underlying clot (arrow, C). D: Laminated, detrital type 3 carbonaceous matter, partly rippled (black arrow), with sedimented clot (white arrow). E: Type 2 clots, whose irregular shape suggests in-situ growth, co-occur with sedimented carbonaceous matter (white arrow). F: Detrital fragment of well-preserved phototrophic biofilm in facies A. G: Phototrophic laminae are generally poorly preserved in the coarse sands of facies A (and facies D).

rophic clots in the Josefsdal Chert are directly related to paleoenvironmental conditions (i.e., distribution of its varied sedimentary facies associations) and the influence of contemporaneous hydrothermal activity (Fig. 2) (see Fig. DR8 for a sketch of the interpreted paleoenvironmental scenario). For example, type 1 biofilms occur on bedding surfaces of sandy sediments across all facies and are relatively common, although not well developed unless in the vicinity of paleohydrothermal activity, where they intercalate with chemotrophic clots (Fig. 3A; Figs. DR6 and DR7A). Likewise, chemotrophic clots occur sporadically throughout the finer-grained sediments, but are only well developed near sites of paleohydrothermal activity (Fig. 3A; Figs. DR6 and DR7A).

Preservation of the interpreted diverse phototrophic and/or chemotrophic communities is, in places, excellent owing to an abundant supply of silica from hydrothermal fluids and Si-enriched seawater and rapid mineralization (e.g., Figs. 3A–3C). However, phototrophic films naturally undergoing varying amounts of degradation (cf. Guido et al., 2010) in high-energy Josefsdal paleoenvironments suffered the taphonomic effects of physical fragmentation, degradation, desiccation, and erosion before preservation (Figs. 3F and 3G; Fig. DR3).

## CONCLUSIONS

In summary, the Josefsdal Chert records a coastal, volcanogenic sedimentary environment in which diverse populations of phototrophic, chemo-organotrophic, and chemolithotrophic microorganisms coexisted. In this interacting microbial-sediment system, microbial colonies were well developed in the vicinity of widespread hydrothermal activity that enhanced high biomass production and, at the same time, enabled extensive biosignature preservation. The ubiquitous evidence for hydrothermal influence suggests that life in the early to mid-Archean Josefsdal setting was thermophilic and that chemotrophs, fuelled by hydrothermal activity, were as common as phototrophs.

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### Archean (3.33 Ga) microbe-sediment systems were diverse and flourished in a hydrothermal context

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