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

Frances Westall1*, Kathleen A. Campbell1,2,3, Jean Gabriel Bréhéret4, Frédéric Foucher1, Pascale Gautret5, Axelle Hubert1,6, Stéphanie Sorieul7, Nathalie Grassineau8, and Diego M. Guido9

1Centre de Biophysique Moléculaire-CNRS, Rue Charles Sadron, 45071 Orléans, France
2LE STUDIUM Institute for Advanced Studies, 1 rue Dupanloup, 45000 Orléans, France
3School of Environment, The University of Auckland, Private Bag 92019, Auckland 1142, New Zealand
4Géo-hydro-systèmes continentaux (GEHCO), Parc de Grandmont 37200 Tours, France
5Université d’Orléans, ISTO, UMR 7327, 45071 Orléans, France and CNRS, ISTO, UMR 7327, 45071 Orléans, France and Bureau de Recherches Géologiques et Minières, ISTO, UMR 7327, BP 36009, 45060 Orléans, France
6Institut des Sciences de la Terre, 1381 rue de la Piscine, 38400 Saint-Martin d’Hères, France
7AIFIRA, Centre d’Etudes Nucléaires de Bordeaux Gradignan, 19 Chemin du Solarium, CS 10120, 33175 Gradignan Cedex, France
8Earth Sciences Department, Royal Holloway University of London, Egham, Surrey TW20 0EX, UK
9CONICET-UNLP, Instituto de Recursos Minerales, Calle 64 Esquina 120, La Plata (1900), Argentina
*E-mail: frances.westall@cnrs-orleans.fr

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 multi-species 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), makes 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, b). On the other hand, no previous investigations have recognized and addressed the syngenetic diversity of Archean microbial paleocommunities—both phototrophs and chemotrophs —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 as both 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 (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 elemental mapping was undertaken on 60–80-µm-thick rock sections by proton induced X-ray emission (PIXE) spectrometry at the Applications Interdisciplinaires des Faiseaux d’Ions en Région Aquitaine (AIFIRA) facility, Centre des Etudes Nucléaires à Bordeaux-Gradignan, France. In-situ δ13C and δ34S measurements were made on polished rock
surfaces and thin sections using a Cameca 1280 HR microprobe at the Le Centre de Recherches Pétrographiques et Géochimiques, Nancy, France. Details of the operating conditions for the various analytical techniques can be found in the Methods section of the GSA Data Repository1.

**SHALLOW-WATER SEDIMENTARY CONTEXT**

The Early Archean Josefsdal Chert comprises a thin package (6–30 m) of silicified, mainly sandy 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 paleo-environment (Fig. 2). The basal portion (Unit 1) comprises swaley to hummocky, cross-stratified, well-sorted sandstone (SCS-HCS) representing upper offshore to shoreface storm deposits.
Figure 2. Stratigraphy and facies associations of the Josefsdal Chert, South Africa.
A: Stratigraphic column (based on outcrops L–M) comprising 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. Unit 1 is dominated by swaley and hummocky cross-stratification (SCS-HCS) (Facies A) (shown in field photograph, B), deposited by storms in the upper offshore to shoreface, with abundant, penecontemporaneous, hydrothermal white to translucent, vertical chert dikes (vcd) and thin chert sills. Unit 2 is dominated by iron-stained, poorly sorted sediments of alternating Facies A and B, with the latter shown in C and interpreted to have been deposited in a shoreface setting that was periodically tidally influenced. Hydrothermal black and white banded chert (D) comprises Facies C, which caps Unit 2. Unit 3 consists of planar laminated volcanic accretionary lapilli and ash (Facies D; shown in E; cf. ref. Lowe, 1999) that was 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 constitutes alternations of Facies A (with rare dessication cracks), C, and D, that accumulated in shoreface to foreshore settings. Biosignatures occur ubiquitously in all facies but are generally infrequent but dense in the vicinity of hydrothermal activity.
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), SCS-HCS (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 (Figs. 1C; DR1). 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 paleo-hydrothermal 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$_2$) and the immediately underlying basalts was due to 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), the Mud of Queensland (MUQ)-normalized (Bohlar et al., 2005) REE + Y patterns and the 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 the 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 intraformational 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. 1B, 1D, and DR5A), sometimes 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 fully lithified, lower (Facies A only) Josefsdal sediments (Fig. 2B). At Josefsdal, our results are consistent with
Evidence for hydrothermal activity driven by the circulation of Si-enriched seawater through cooling lavas elsewhere in the Barberton Greenstone Belt (Hofmann and Harris, 2008) and the Pilbara (e.g. van den Boorn et al., 2007).

**Figure 3.** Carbonaceous material types and characteristics in the Josefsdal Chert, South Africa. A: Type 1, dark wavy, phototrophic laminae (black arrows), entrap and are underlain by dark, chemotrophic clots (Type 2; selected clots labeled as C), and embedded in a clear chert matrix (Facies C). B,C: Details of Type 1 phototrophic layer, showing fine, undulating laminae entrapping chert matrix (Facies C). B,C: Details of Type 1 phototrophic layer, showing fine, undulating laminae entrapping detrital grains (arrows, b) and compensation of laminae over underlying Type 2 clot (arrow, c). D: Detrital Type 2 clot (white arrow) sedimented together with fine-grained, detrital Type 3 carbonaceous matter forming very fine laminae (e.g., black arrow at ripple cross-lamina) (Facies C). E: Type 2 clots with irregular, spiky protuberances suggest in-situ growth, possibly in a silica gel (clear matrix). White arrows mark discontinuous laminae of Type 3 carbonaceous matter (Facies C). F: Transported, silicified fragment of inferred Type 1 phototrophic biofilm (Facies A). G: Thin horizons of relatively poorly preserved (degraded), wavy, Type 1 phototrophic laminae in coarse sand of Facies A (also observed in Facies D).

**IN-SITU MICROBIAL COMMUNITIES AND DETRITAL CARBON**

In the Archean, 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 to a few tens of micrometers) that often 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 in 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^{13}C_{VPDB} (Vienna Peedee belemnite) are consistent with the Type 1 films representing microbial mats utilizing a phototrophic metabolism (c.f. Tice, 2009). The previously reported association of chemo-organotrophic sulfate-reducing bacteria (SRB) with one of these biofilms (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 biofilm in question of δ^{13}C_{VPDB} signatures as light as −45‰ and depleted δ^{34}S_{CDT} (Canyon Diablo Troilite) values of −24‰ (c.f. 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 (c.f. Bailey et al., 2009) formed of carbon-coated, sand-sized (50–500 µm) volcanic clasts (Figs. 3A; 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 (spiky protuberances or cauliflower-shaped), 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 modern, hydrothermally-influenced, basaltic sediments (Campbell et al., 2001; Callac et al., 2013). High concentrations of clots at sites of greatest hydrothermal activity (Figs. 3A and DR7A) suggests thermal fluids as a principal energy source, providing inorganic (e.g. H_2, CO_2) 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 (cf. 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 chemotrophic 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 paleo-hydrothermal activity, where they intercalate with chemotrophic clots (Figs. 3A, DR6 and DR7A). Likewise, chemotrophic clots occur sporadically throughout the finer grained sediments, but are only well-developed near sites of paleo-hydrothermal activity (Figs. 3A, DR6 and DR7A).

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

CONCLUSIONS

In summary, the Josefsdal Chert records a coastal, volcanogenic sedimentary environment in which diverse colonies of phototrophic, chemo-organotrophic, and chemolithotrophic microorganisms co-existed. 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-Mid Archean Josefsdal setting was thermophilic and that chemotrophs, fuelled by hydrothermal activity, were as common as phototrophs.

ACKNOWLEDGMENTS

We are grateful to B. Cavalazzi (University of Bologna) and J. Torres (CNRS-CBM) for assistance with field work (2012); S. Janiec (CNRS-ISTO) for making the thin sections; E. Deloule and N. Bouden (CRPG) for help with the microprobe analyses; E. Biguereau, D. Bouvard and C.P. Carry (University of Grenoble) for preparation of the C and S isotope standards; C. Ramboz for discussions regarding
hydrothermal signatures; C. Rollion-Bard for preliminary in-situ C isotope tests; and L.M. Cotterall (University of Auckland) for drafting several figures. This work was supported by grants to Westall from ANR-09-BLAN-0219-01, CNRS-MI-2014, and the MASE project, supported by the European Community’s Seventh Framework Programme (FP7/2007-2013) under Grant Agreement number 607297; to Campbell from the Marsden Fund (RSNZ); to Guido and Campbell from the National Geographic Society; and to Campbell from a LE STUDIUM Institute for Advanced Studies research fellowship.

REFERENCES CITED


