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Carbon isotopic analyses of ca. 3.0 Ga microstructures imply planktonic autotrophs inhabited Earth’s early oceans

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ABSTRACT

The ca. 3 Ga Farrel Quartzite (FQ, Western Australia) contains possible organic microfossils of unusual spindle-like morphology that are surprisingly large and complex, preserved along with spheroids. The unusual nature of the possible fossils, coupled with their antiquity, makes their interpretation as biogenic difficult and debatable. Here, we report 32 in situ carbon isotopic analyses of 15 individual FQ specimens. The spheroids and the spindle-like forms have a weighted mean δ13C value of −37‰, an isotopic composition that is quite consistent with a biogenic origin. Both the spheroids and the spindle-like structures are isotopically distinct from the background organic matter in the same thin section (weighted mean δ13C value of −33‰), which shows that the preserved microstructures are not pseudofossils formed from physical reprocessing of the bulk sedimentary organic material. When considered along with published morphological and chemical studies, these results indicate that the FQ microstructures are bona fide microfossils, and support the interpretation that the spindles were planktonic. Our results also provide metabolic constraints that imply most of these preserved microorganisms were autotrophic. The existence of similar spindles in the ca. 3.4 Ga Strelley Pool Formation of Australia and the ca. 3.4 Ga Onverwacht Group of South Africa suggests that the spindle-containing microbiota may be one of the oldest, morphologically preserved examples of life. If this is the case, then the FQ structures represent the remains of a cosmopolitan biological experiment that appears to have lasted for several hundred million years, starting in the Paleoarchean.

INTRODUCTION

Interpretation of microstructures in the ca. 3.0 Ga Farrel Quartzite (FQ, Western Australia) has been challenging because of the large size and unusual morphology of spindle-like forms (Sugitani et al., 2007, 2009b). Morphologies of microfossils preserved in chert along with the δ13C composition of bulk organic matter (OM) have provided evidence for and insights into the metabolic capabilities of Precambrian life (Oehler et al., 1972; Schidlowski, 2001; Schopf, 1994, 2006). However, interpretations from bulk isotopic analyses are limited, as the carbon could derive from indigenous organisms, reworked OM, abiotic organics, and post-depositional contaminants. Ideally, δ13C values should be obtained for individual microfossils, and work has shown that reliable δ13C values of individual Proterozoic microfossils can be obtained with secondary ion mass spectrometry (SIMS; House et al., 2000).

Attempts to extend such fossil-specific data to the Archean, however, have proven difficult due to low count rates from microfossil-like structures of that antiquity (D.Z. Oehler and C.H. House, unpublished data). Accordingly, δ13C studies of Archean OM forms are limited to values published for carbonaceous filaments from Archean-aged siliceous veins (Ueno et al., 2001), δ13C values obtained for a number of organic spots in Strelley Pool Formation thin sections (Wacey et al., 2011), and recent measurements of a suite of samples of Neoarchean sedimentary OM (Wil- liford et al., 2011). Importantly, prior to the work reported here, there have been no published in situ Archean analyses that reliably demonstrate an isotopic distinction between specific microstructures and background OM in the rock, thus providing information that is unique from published whole-rock data. Here, we present δ13C analyses of multiple examples of individual microstructures. The data show a clear difference between the δ13C values of the structures and background OM, providing new insight into the evolutionary significance of the targets.

The Farrel Quartzite, in the northeastern part of the Pilbara Craton (Fig. DR1 in the GSA Data Repository; Hickman, 2008), provides an intriguing assemblage of organic microstructures for study because cherts in this deposit contain multiple, moderately well-preserved specimens composed of relatively dense OM. Key constituents of this assemblage have been previously studied in detail (Sugitani et al., 2007, 2009a, 2009b), such that their morphological variability, taphonomic attributes, and population distributions are known. However, the spindle-like forms in the FQ seem surprisingly large (~20–60 μm in length) for such ancient microorganisms, though Javaux et al. (2010) have recently reported 3.2 Ga, ~300 μm, organic spheroids that they interpret as microfossils living in the photic zone. Nevertheless, the FQ spindles are additionally of uncertain biogenicity because of their morphology, which resembles possible artifacts produced by OM that could have accumulated in and around pre-existing crystals. Thus, the acquisition of in situ δ13C data from the FQ microstructures can, as called for by Wacey (2009), test the biogenicity of the spindle-like structures, as well as other constituents of the assemblage. Moreover, these data have the potential to provide an isotopic constraint on metabolisms represented in this deposit. Here, we report the results of 46 in situ carbon isotopic analyses of the ca. 3.0 Ga FQ, obtained using SIMS, including 32 analyses on 15 individual microstructures.

METHODS

Three polished thin sections from the Mount Grant locality (Fig. DR1; Sugitani et al., 2007) were assessed for microstructures appropriate for SIMS analysis. Microstructures at the surface and with a high quality of preservation were selected. All structures selected were within the thin section GFSV3-DO1. δ13C compositions were determined with the University of California–Los Angeles (UCLA) Cameca 1270 using a multicollector configuration with 13C–detected by an off-axis electron multiplier (EM) and 13C/12C–measured using the central EM. The molecular ion

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GSA Data Repository item 2013177, Figures DR1–DR5 and Table DR1, is available online at www.geosociety.org/pubs/ft2013.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

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produces a stronger count rate than $-^{14}$C and its mass permits the on-axis detector to be used, allowing for verification of targets by ion imaging prior to analysis. SIMS was performed using a 0.01–0.5 nA, ~15 μm, Cs$^+$ primary beam (with lower intensities required for spindles and the full range required for the other analyses). For analysis of background OM, the primary ion beam was occasionally moved off of a target to an area showing diffuse or irregular OM that were not obvious fragments of the microstructure.

Charge compensation was achieved using a normal incident electron gun and a gold coat. The analyses were calibrated against repeated analyses of PPRG#215–1 Precambrian chert, as previously used for Bitter Springs and Gunflint microfossils (House et al., 2000; Williford et al., 2013). The instrumental mass fractionation was found to be similar to past experience with these conditions and quasi-simultaneous arrival effects were estimated to be unimportant (see notes from Table DR1 in the Data Repository).

**RESULTS**

Twenty-one (21) SIMS $\delta^{13}$C$_{\text{PDB}}$ analyses were obtained on nine specimens of spheroids (Fig. 1; Fig. DR2) that occur within clusters in the FQ; this group had $\delta^{13}$C$_{\text{PDB}}$ values ranging from $-31.8\%e \pm 1.4\%e$ to $-44.2\%e \pm 1.3\%e$ (weighted mean = $-36.9\%e \pm 0.3\%e$; mean square weighted deviation [MSWD] = 6.8; standard deviation [SD] = 3.5). In addition, ten SIMS $\delta^{13}$C$_{\text{PDB}}$ analyses were obtained on six specimens of the spindle-like microstructures (Fig. 2; Fig. DR2); these structures had $\delta^{13}$C values ranging from $-35.6\%e \pm 1.4\%e$ to $-40.5\%e \pm 1.2\%e$ (weighted mean = $-37.0\%e \pm 0.4\%e$; MSWD = 1.5; SD = 1.6). Finally, 14 analyses were obtained of background OM (weighted mean = $-32.7\%e \pm 0.6\%e$, MSWD = 1.6; SD = 3.3). The complete data set is shown in Figure 3 and in Table DR1.

The results show that the mean $\delta^{13}$C value for the spindles is similar to that for the spheroids and that both the spindles and spheroids are significantly depleted in $^{13}$C compared to background OM. In addition, while the mean isotopic compositions for the spindles and spheroids are similar, the range of values for the spindles is considerably narrower than that for spheroids, which has a SD more than twice as large. While the values reported here for background OM appear to vary greatly, much of this variance is attributable to limited counts as indicated by the MSWD (or reduced $\chi^2$) value which is close to unity showing that the variance observed is consistent with that expected by the uncertainties of the analyses.

**DISCUSSION**

$\delta^{13}$C Values for Spheroids and Spindles

Our mean SIMS $\delta^{13}$C value for background OM ($-32.7\%e \pm 0.6\%e$) in the thin section matches published whole-rock values of total organic carbon in the FQ ($-33.2\%e$, SD = 0.8; Sugahara et al., 2010; Sugitani et al., 2007). This correspondence of values from two disparate methods

![Figure 1. Clusters of spheroidal microfossils of the Farrel Quartzite (Western Australia) in transmitted optical light (A, C, E, and G) and in reflected light (B, D, F, and H). Reflected light images show locations and secondary ion mass spectrometry (SIMS)–measured $\delta^{13}$C values in per mil ($\%e \pm 1\sigma$) of analyzed organic matter at the surface. Isotope data are listed in Table DR1 (see footnote 1). Cluster identifications (in Table DR1) are as follows: A and B, FQ1_11; C and D, FQ2_5; E and F, FQ2_8; G and H, FQ2_9. Actual locations of SIMS analyses are shown by dashed ovals. Locations were determined after SIMS was completed by optical microscopy. Figure DR3 (see footnote 1) shows examples of SIMS analysis pits.](image1)

![Figure 2. Spindle-like microstructures of the Farrel Quartzite (Western Australia) in transmitted optical light (A, C, and E) and in reflected light (B, D, and F). Reflected light images show locations and secondary ion mass spectrometry (SIMS)–measured $\delta^{13}$C values in per mil ($\%e \pm 1\sigma$) of analyzed organic matter at the surface. Isotope data are listed in Table DR1 (see footnote 1). Spindle identifications (in Table DR1) are as follows: A and B, FQ2_5; C and D, FQ2_6; E and F, FQ1_4. Actual locations of SIMS analyses are shown by dashed ovals. Locations were determined after SIMS was completed by optical microscopy. Figure DR4 (see footnote 1) shows examples of SIMS analysis pits.](image2)
provides verification of our results, as well as the way we identified background OM. Almost certainly some of the background OM is derived from degraded microstructures, and such an origin might be reflected by our first two OM analyses (−38.3‰ ± 2.3‰ and −40.3‰ ± 5.3‰), which were taken near the spindle chain (sample FQ1_4 in Table DR1). However, our SIMS results reported here are generally consistent with an OM isotopic composition of about −33‰, with a MSDW for all analyses at 1.6 (where a value of unity indicates that variance is accounted for by the uncertainties of the analyses). Further, the weighted mean of our four most precise OM measurements (all with uncertainties better than ±1.9‰) is −33.1‰ ± 0.7‰, and one background OM measurement (FQ2_7_uni1_bk) gave a strong carbon signal, like a microstructure, and still showed a value (−31.8‰ ± 1.4‰) consistent with the other background OM. Our results, thus, clearly demonstrate that the bulk of the OM in the rock is isotopically distinct from the preserved spindle and spheroid microstructures (P-values < 0.01). These results argue against the possibility that the spindles are pseudofossils created by accumulation of sedimentary OM around, within, or between minerals, as if that were the case, then it would be expected that the isotopic compositions of the spindles would mimic that of the background OM. Our data also indicate that the spindle and spheroid microstructures make up a small fraction of the total organic carbon preserved. The QF cherts have been interpreted to represent a coastal depositional setting with possible nonmarine hydrological influence and a lack of high-temperature hydrothermal input (Sugahara et al., 2010). This model is consistent with the preserved OM being derived from a mixture of planktonic and local sources. Possibly related, δ13C values for deepwater OM in sediments from the younger ca. 2.7 Ga Hamersley Basin are ~10‰ more 13C-depleted than values for shallow-water OM (Eibenrode and Freeman, 2006).

Based on the observation that the carbon in the FQ spheroids (with a mean δ13C composition of −36.9‰ ± 0.3‰) are more 13C-depleted than background carbon (mean δ13C = −32.7‰ ± 0.6‰), it is unlikely that the spheroids represent fermenting or respiring heterotrophs that have consumed background carbon. Such metabolism would likely generate cells isotopically heavier than their substrates (Coffin et al., 1989; Hall et al., 1999; Macko and Estep, 1984) through expulsion of 13C-depleted carbon by dissimilatory metabolisms. For this reason, we interpret the results as suggesting that the spheroids represent autotrophic cells. However, methanogenic or methanotrophic pathways cannot be ruled out by the data, as these metabolisms can result in diverse δ13C compositions (Conrad, 2005; House et al., 2009; Londry et al., 2008). Accordingly, the wide range of δ13C values for the spheroids could reflect an element of methane incorporation, perhaps accounting for some of the lightest values.

As noted, both the spindles and spheroids have mean δ13C values of −37‰ (Fig. 3). A general explanation of these light values would be that CO2 levels in the environment where the organisms grew were high, resulting in maximal fractionation between autotrophic microbes and their source CO2. The results are broadly consistent with carbon fixation via either the Calvin cycle or acetyl-CoA pathway (and not the 3-hydroxypropionate cycle or reductive tricarboxylic acid cycle [Zerkle et al., 2005]). Such relatively light isotopic values may reflect a planktonic lifestyle where CO2 would not be limiting as it can be in uncirculated benthic habitats (Kaufman and Xiao, 2003). This possibility is supported by morphological attributes of the spindles that suggest a planktonic habit (Sugitani et al., 2007; Oehler et al., 2010) including their flanged lenticular shape, occasional internal vacuole-like structures, and the possible association with small non-clustered spheroids (Fig. DR5). Other OM in the chert would likely have been produced at or near the site of deposition from microorganisms with less-resilient cell walls. The highly 13C-depleted values of some of the spheroids might represent some incorporation of previously fractionated carbon, such as methane, as noted above, but such an explanation seems less likely for the spindles than the spheroids, given their narrow isotopic range, right at the lower limit expected for the Calvin cycle (Fig. 3).

Consideration of Published Raman and NanoSIMS Results

These microstructures have also been characterized by Raman spectroscopy and NanoSIMS. Raman results showed that all the microstructures are composed of kerogen with spectral characteristics indistinguishable from that of the background OM, suggesting that both structures and background OM are syngenetic with deposition (Schopf et al., 2005; Sugitani et al., 2007; J.W. Schopf, 2012, personal commun.). The NanoSIMS analyses showed that microstructures have carbon, nitrogen, and sulfur distributions similar to elemental distributions of undisputed, younger microfossils (Oehler et al., 2010) including their flanged lenticular shape, logical attributes of the spindles that suggest a planktonic habit (Sugitani et al., 2007; Oehler et al., 2010). In addition, the NanoSIMS element distributions for the spindles and spheroids were similar to each other. Moreover, the data from the spindles showed that material forming their exteriors is identical to that which fills many of the specimens, arguing against the spindle-like morphology being a result of accumulation of kerogen on the exteriors of minerals. This conclusion parallels our conclusion here that the spindles are not pseudofossils made up of background OM.

CONCLUSIONS

The δ13C values reported here, along with previous morphological, Raman, and NanoSIMS analyses, provide a strong argument that FQ microstructures represent a ca. 3 Ga, biologically diverse, microbiota. Further, the tight clustering of data for spindles at values distinctly more 13C-depleted than the background OM can be explained if the microorganisms now preserved as spindles (and potentially spheroids) were planktonic, allowing them to grow without CO2 limitation and thus exhibit maximal isotopic fractionation for their biomass (Kaufman and Xiao, 2003). This possibility is supported by morphological attributes of the spindles that suggest a planktonic habit (e.g., Oehler et al., 2010). The spindles and spheroids are indistinguishable from structures found in the older, ca. 3.4 Ga Strelley Pool Formation (Sugitani et al., 2010, 2013); the spindles are also similar to structures reported by Walsh (1992) from the ca. 3.4 Ga Onverwacht Group (de Ronde and de Wit, 1994; Lowe and Byerly, 2007). The existence of similar microfossils in disparate locations...
at ca. 3.4 Ga suggests that this unusual microbiota extends back to at least that earlier age, and was long lived and geographically widespread. Such a cosmopolitan distribution might well reflect a planktonic habit for these microorganisms. That habit, in the Earth’s primeval oceans, was likely to have been relatively unchallenged, perhaps contributing to the sustained success of this unusual paleobiota.

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