



Short communication

Identification of an Archean marine oxygen oasis

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ABSTRACT

The early Earth was essentially anoxic. A number of indicators suggest the presence of oxygenic photosynthesis ~2700–3000 million years (Ma) ago, but direct evidence for molecular oxygen (O₂) in seawater has remained elusive. Here we report rare earth element (REE) analyses of ~2800 million year old shallow-marine limestones and deep-water iron-rich sediments at Steep Rock Lake, Canada. These show that the seawater from which extensive shallow-water limestones precipitated was oxygenated, whereas the adjacent deeper waters where iron-rich sediments formed were not. We propose that oxygen promoted limestone precipitation by oxidative removal of dissolved ferrous iron species, Fe(II), to insoluble Fe(III) oxyhydroxide, and estimate that at least 10.25 μM oxygen concentration in seawater was required to accomplish this at Steep Rock. This agrees with the hypothesis that an ample supply of dissolved Fe(II) in Archean oceans would have hindered limestone formation. There is no direct evidence for the oxygen source at Steep Rock, but organic carbon isotope values and diverse stromatolites in the limestones suggest the presence of cyanobacteria. Our findings support the view that during the Archean significant oxygen levels first developed in protected nutrient-rich shallow marine habitats. They indicate that these environments were spatially restricted, transient, and promoted limestone precipitation. If Archean marine limestones in general reflect localized oxygenic removal of dissolved iron at the margins of otherwise anoxic iron-rich seas, then early oxygen oases are less elusive than has been assumed.

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1. Introduction

Progressive oxygenation of the atmosphere and hydrosphere had a major long-term effect on Earth-surface environments and the development of life. Its inception is widely attributed to the development of oxygenic photosynthesis by cyanobacteria during the Archean (Canfield, 2005; Holland, 2006). The existence of oxygenic photosynthesis ~2700 to 3000 Ma ago (Canfield, 2005; Nisbet et al., 2007; Buick, 2008; Schwartzman et al., 2008; Schopf, 2011) is suggested by chromium isotopes and redox-sensitive metals in 2920–2980 Ma sediments (Crowe et al., 2013), very negative carbon isotope values 2760 Ma ago that suggest methane oxidation (Hayes, 1994), phototrophic filaments in 2720 Ma stromatolites (Buick, 1992), hydrocarbon biomarkers for oxygenic photosynthetic cyanobacteria at 2720–2560 Ma (Eigenbrode et al., 2008), and correlated iron and molybdenum isotope compositions in 2680 Ma sediments (Czaja et al., 2012). Much of the early free oxygen (O₂) is thought to have been removed by reaction

with dissolved iron in seawater, contributing to widespread deposition of deep water iron- and silica-rich banded iron formation (Canfield, 2005; Holland, 2006). Nonetheless, substantial amounts of molecular oxygen might have accumulated locally in protected shallow-water environments that favored cyanobacterial productivity (MacGregor, 1927; Cloud, 1965; Kasting, 1992) and were sufficiently isolated so that the oxygen was not all immediately scavenged (Hayes, 1983). With a low rate of atmospheric exchange, O₂ levels in these ‘oxygen oases’ (Fischer, 1965) could have approached 0.016 atm (0.08 PAL), even under an anoxic atmosphere (Kasting, 1992; Pavlov and Kasting, 2002; Olson et al., 2013).

2. Steep Rock

We collected samples from ~2800 Ma (Fralick et al., 2008) limestones at Steep Rock Lake, 5 km north of Atikokan, Northwestern Ontario, in the central Wabigoon Subprovince of the Canadian Shield (Supplemental Data). This 500 m thick (Fig. 1) unit of calcium carbonate (CaCO₃) was chosen for study as the biogenic structures are well preserved (Wilks and Nisbet, 1988) and there is little evidence of chemical alteration (Veizer et al., 1982). The succession was deposited on an oceanic, volcanic plateau (Fralick et al., 2008)

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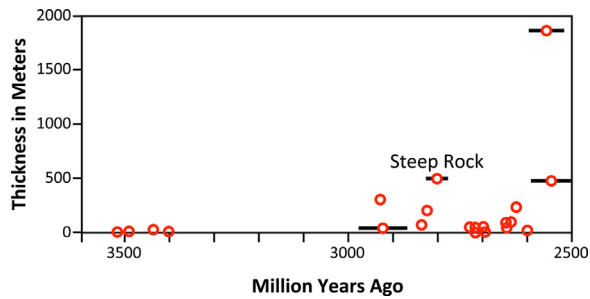


Fig. 1. Thickness-age plot of sedimentary calcium and magnesium carbonate rock units older than 2500 Ma (see Supplemental Data for details). Age error bars are horizontal.

and is laterally and vertically transitional to iron formation (Fig. 2). It overlies ~3000 Ma tonalitic gneiss eroded by a paleo-channel network back-filled with fluvial sandstones and conglomerates (Wilks and Nisbet, 1988; Fralick et al., 2008). These were covered by a transgressing sea that deposited dominantly CaCO_3 , with thin layers of dolomite [$\text{CaMg}(\text{CO}_3)_2$], ankerite [$\text{Ca}(\text{Fe},\text{Mg}(\text{CO}_3)_2$), and siderite (FeCO_3) that variously formed as primary, and as early and late diagenetic, products. The Steep Rock carbonate deposit therefore occupied a shallow platform environment adjacent to deeper water. Various morphologies, including columnar and domical forms, of relatively small (<0.5 m thick) crinkly laminated stromatolites that closely resemble lithified microbial mats occur in the lower part of the limestone (Wilks and Nisbet, 1985), and the upper part is dominated by large domes up to 5 m across composed of interlayered fenestral fabrics, interpreted as microbial, and crystal fan layers, that formed as aragonite seafloor precipitates (Sumner and Grotzinger, 2000) (Fig. 3). Manganese- and iron-oxide deposits ('Paint Rock') directly overlie the limestone (Wilks and Nisbet, 1988; Fralick et al., 2008). Steep Rock limestone formation was terminated as relative sea level rose and the platform was covered by these deeper water manganese and iron deposits (Fralick et al., 2008).

3. Methods

Nine samples of iron formation and 23 samples of limestone were selected to be representative of the succession and

lithotypes present. Each sample was cut from a single fabric component, then hand crushed and as much of the white blocky cement as possible was removed before final crushing. The samples were analyzed for rare earth elements (REE) by inductively coupled plasma mass spectrometry (ICP-MS) at the Instrumentation Laboratory at Lakehead University. Calcium was analyzed by ICP-AES at the same facility using the same sample preparation techniques. For these analyses, 500 mg aliquots of powder were digested by a five-stage procedure involving initial release of the volatile phase using dilute hydrochloric acid, followed by three separate treatments with nitric and hydrofluoric acid separated by periods of drying. Finally, the sample was dissolved in dilute nitric acid and diluted 1000 times. Strontium was analyzed at the Geoscience Laboratories of the Ontario Ministry of Northern Development and Mines in Sudbury, Ontario. Two hundred milligram aliquots of powder were digested by a two-stage procedure involving initial decomposition in a closed beaker by a mixture of HF with lesser amounts of HCl and HClO_4 followed by a second mixture of dilute HCl and HClO_4 . For REE, Sr and Ca analyses, blanks and appropriate standards were run with the samples. Accuracy and precision were evaluated by analyzing duplicate samples and the standard reference materials imbedded in the run, and were within acceptable limits. The REE data were standardized to post-Archean Australian Shale. The nine samples of iron formation showed no significant difference in REE pattern shape between the various sample lithologies. A limestone sample from the base of the formation contained sufficient siliciclastic material for this component to dominate its REE pattern, and it is plotted as a siliciclastic with two shale samples from the iron formation (Fig. 4).

To validate the presence of negative Ce anomalies we conducted sample treatment, analyzed comparative samples, and analyzed samples using a different laboratory, as follows. Two samples (SR22A, SR16A) were selected for different analytical treatment. A partial digestion using 5% acetic acid was performed on sample SR22A, followed by desiccation of the liquid, dissolution in 2% nitric acid and analysis of REEs on an ICP-MS at Lakehead University (Fig. 5a). Four samples from the Paleoproterozoic Gunflint Formation, with REE abundances lower than SR22A, were also analyzed. To bring the abundances in solution to well above detection limits, the dilution was limited to fifty times. The four Gunflint samples showed no significant negative Ce anomaly, whereas SR22A had an obvious negative Ce anomaly (Fig. 5a). The REE concentrations

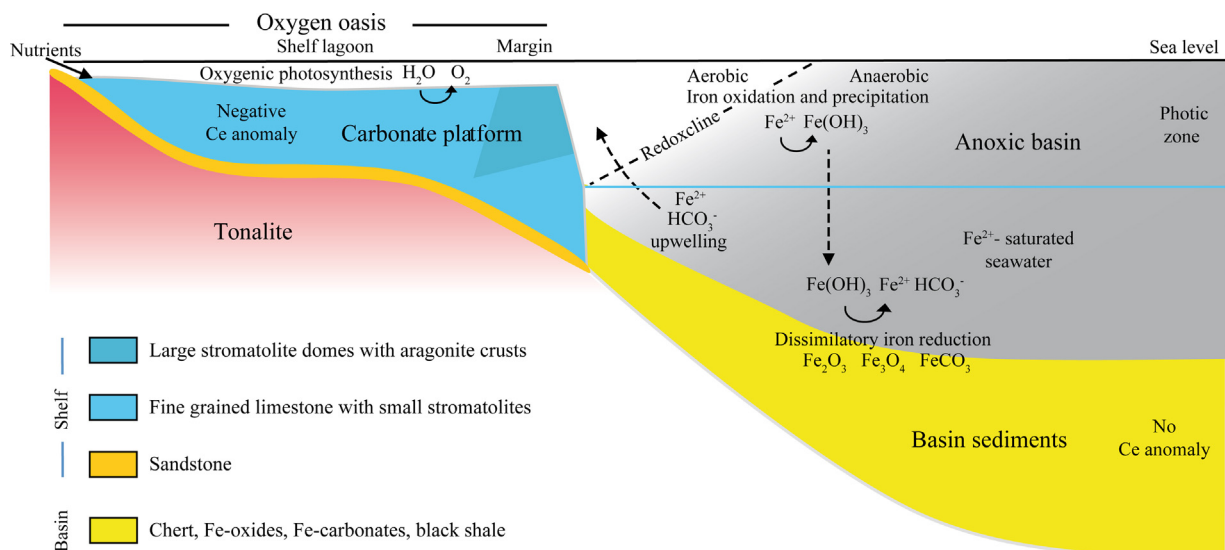


Fig. 2. Interpretive cross-section of Steep Rock oxygenated carbonate platform and adjacent anoxic and iron-rich basin, showing inferred stratigraphic relationships, conditions, and microbial metabolic processes (not to scale).

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