



A biological switch at the ocean surface as a cause of laminations in a Precambrian iron formation



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ABSTRACT

Banded iron formations (BIFs) exhibit alternating silica- and iron-rich laminae, potentially reflecting the dynamics of the paleo-environments in which they were formed, although the exact mechanism remains unclear. Here the formation of a 2.7–2.9 Ga BIF from Dharwar Craton, India, is deciphered by analyzing the inter-band variations of the redox-sensitive isotope biomarkers, $^{15}\text{N}/^{14}\text{N}$ and $^{56}\text{Fe}/^{54}\text{Fe}$. Organic matter with $\delta^{15}\text{N}$ values as high as $+12.0 \pm 0.8\text{‰}$ appears to be trapped in silica. Iron oxides exhibit systematically positive $\delta^{56}\text{Fe}$ values, ranging between $+0.80 \pm 0.05\text{‰}$ and $+1.67 \pm 0.02\text{‰}$. Compared to the iron-rich bands, silica-rich bands, which show higher $\delta^{56}\text{Fe}$ values, exhibit an order of magnitude higher concentrations of ^{15}N -rich organic nitrogen, normalized by the abundances of its host silica. The presence of ^{15}N -rich organic matter may imply the emergence of a modern-like biological nitrogen cycle that requires the formation of oxidized nitrogen compounds. The higher concentration of ^{15}N -rich organic nitrogen for the silica-rich bands possibly suggests that the photosynthetic activity was higher during the formation periods of these bands. The heavier iron isotope compositions of the silica-rich bands cannot be explained alone by iron oxidation through probable pathways. The relative ^{56}Fe -enrichment in silica-rich bands is explained here by the progressive dissolution of iron oxides to the ocean, through iron reduction by ^{15}N -rich organic matter actively produced at the ocean surface. The formation of iron-rich bands possibly corresponds to periods of reduced biological productivity, when precipitated iron was not effectively dissolved to the ocean. The observed shift in the organic concentration between Fe- and Si-rich bands could be the switch that triggered the BIF laminations. This shift could conceivably represent periodic fluctuations in the oxygen generation, which possibly occurred over periods of millennia, at the dawn of the Great Oxidation Event.

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1. Introduction

It is generally accepted that Earth began with a highly reducing surface environment and shifted to an oxidizing one, although the timing of this oxidation is a matter of debate. One major school has proposed that the first global oxidation of the atmosphere occurred at approximately 2.32 Ga (Holland, 2002; Bekker et al., 2004), whereas others have claimed that it occurred much earlier, possibly in the Hadean or early Archean (Ohmoto et al., 2006). The mechanisms triggering this major oxidation event are not well understood. If microorganisms in the early Earth's

ocean generated the oxygen molecule, a thorough understanding of how Earth prepared its surface to sustain these “oxygenators” is lacking.

Banded iron formations (BIFs) bear witness to this environmental shift. Worldwide, BIFs were formed largely between 3.8 to 1.8 Ga, with a few exceptions at ~ 0.7 Ga (Klein, 2005), by the oxidation of iron in the ocean water. However, several key issues must be addressed to understand the formation of BIFs and their link to the oxygenation of the hydrosphere–atmosphere system, as well as to the evolution of the biosphere. (1) How did the ferrous iron dissolved in the earliest anoxic ocean precipitate to the ocean-floor? Many alternatives that explain the oxidation process of iron have been proposed (Beukes, 2004), making it difficult to present a clear-cut logic of how exactly BIFs reflect the oxygenation of the ocean. (2) Why are the iron formations so often laminated?

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The lamination has classically been interpreted as representing the fluctuation in the deposition rate of iron-bearing minerals, caused by geological factors, such as fluctuations in ocean temperatures (Posth et al., 2008) or hydrothermal inputs (Morris, 1993; Steinhöfel et al., 2009; Wang et al., 2009).

Nitrogen and iron isotopes are powerful tools to decipher paleo-environments. However, not many studies combine these indicators to compare their sensitivities to various environmental parameters (Shen et al., 2006; Thomazo et al., 2009). One of the most important roles common to both elements is that of electron acceptor (NO_3^- or Fe^{3+}), substituting molecular oxygen under O_2 -poor conditions. Metabolic processes involving these elements are known to produce distinct isotopic fractionations (Dauphas and Rouxel, 2006; Brandes et al., 2007; Sigman et al., 2009; Pinti and Hashizume, 2011). Prominent ^{15}N -rich isotope signatures are observed among BIF samples from the Neoproterozoic or Paleoproterozoic ages, the periods that correspond to the dawn of the Great Oxidation Event (Thomazo et al., 2009). Based on this observation, a detailed nitrogen isotope study was performed at these ages in the current work, since the ^{15}N enrichment was proposed to be associated with the increase in the oxygen concentration in the ocean (Shen et al., 2006; Godfrey and Falkowski, 2009). Because the causal link between the oxidation/precipitation of iron and the oxidation of the ocean is hotly debated (Beukes, 2004), we have explored the correlation between nitrogen and iron isotopes in the BIF sample.

In this study, intensive analyses were carried out on individual bands from a single BIF sample, to determine a novel relationship between N and Fe isotope compositions. A single location was chosen and held constant, and the shift in information across bands was carefully interpreted in order to reconstruct the dynamics of the paleo-environments, particularly the redox condition of the paleo-ocean. Such information would be difficult to obtain by comparing analytical data from samples at different locations, since they would naturally reflect different environmental factors specific to their geological settings.

The selected BIF sample, belonging to the Neoproterozoic era, was previously well studied from both mineralogical and geological perspectives (Orberger et al., 2012; Jayananda et al., 2013, 2015). It was chosen based on the following criteria: (1) The sample records low grade metamorphism, which is an important criterion for the preservation of the primordial nitrogen isotope information. (2) The sample records minimal contribution from crustal sources. Crustal materials may contain abundant nitrogen in the form of ammonium ions, which could mask the primordial nitrogen isotopic information carried by organic matter.

Isotope compositions of three elements (Fe, N and Ar) from ten different iron-oxide or silica bands from the selected BIF sample were analyzed, along with the abundances of sixty elements (C, N, Ar, and 57 solid elements). Elements and isotopes other than those of the main proxies (N and Fe) not only helped to support key discussions of the two isotope systems, but also served in discriminating the heavily altered bands, indispensable to ensuring the veracity of the paleo-environmental constraints derived from the analyses of such ancient rock samples.

2. Materials and methods

2.1. Geology and sample descriptions

The Dharwar craton of southern India (Fig. 1) preserves a large section of Paleoproterozoic to Neoproterozoic continental crust that contain vast areas of 3.4–3.0 Ga TTG-type gneisses, two generations of volcanic-sedimentary greenstone sequences, the older Sargur Group and the younger Dharwar Supergroup, and 2.62 Ga and 2.56–2.52 Ga potassic to sanukitoid intrusions (Jayananda et

al., 2013, 2015). Among the greenstone sequences, the Sargur Group comprises komatiite–tholeiite volcanics with interlayered sediments including quartzite, pelites, carbonates and BIFs. The Dharwar Supergroup is further sub-divided into the lower Bababudan Group and the upper Chitradurga Group. The Bababudan Group (2.91–2.72 Ga) contains shallow-water platform sedimentary sequences, such as oligomict conglomerate/quartzite, followed by mafic to felsic lava flows and thick BIFs, whilst the Chitradurga Group (2.7–2.65 Ga) comprises of polymict conglomerate, basaltic-felsic lava flows, and argillite–greywacke–BIFs.

The studied BIF-14a (Fig. 1) belongs to the Bababudan Group, part of the Chitradurga basin of the Western Dharwar craton. The Bababudan Group is affected by low-grade metamorphism corresponding to greenschist to lower amphibolites facies. The time frame of the metamorphism is not well constrained, however, it could be related to the intrusion of the 2.62 Ga Arsikere granite, or the subsequent regional thermal event, which affected the whole Archean crust close to 2.5 Ga (Jayananda et al., 2013).

At the BIF-14a sample location, the lithological succession begins with oligomict conglomerates at the base, overlain by mafic volcanics, and a 500 m-thick BIF comprised of alternate Fe-oxide and silica layers at the highest stratigraphic levels. The primary structure of the volcanics and the sediments are well preserved, although the rocks experienced lower greenschist facies conditions. The studied Bababudan Group is associated with sedimentary facies of continental-shelf environments, which is a characteristic feature of Superior-type iron formations (Bekker et al., 2010).

Sample BIF-14a is composed of millimeter- to centimeter-thick alternating quartz and iron-oxide bands (Fig. 2a). The massive iron-oxide bands consist of euhedral crystals ($\sim 20\ \mu\text{m}$) of magnetite, partially or completely transformed into hematite (martite), which show dissolution features along parting planes. This transformation took place probably during the regional metamorphism mentioned above. The Bababudan Group experienced greenschist facies conditions as shown by the paragenesis of grunerite (Fe–Mg amphibole) + biotite + quartz + magnetite in the studied sample (Orberger et al., 2012). Rare goethite particles are present (Orberger et al., 2014). Micrometric sized Ca–Mg–Fe carbonates and sulfides (galena, pyrite, chalcopyrite), and minor apatite, monazite, and xenotime occur locally as inclusions in or interstitial to quartz.

At the interface of the massive iron-oxide and quartz bands or within the quartz bands, there are iron-oxide alignments a few tens of micrometers thick and individual iron-oxide grains. Some of these iron oxides have spherulitic structures (Orberger et al., 2012). These alignments have been interpreted as cavities formed by the dissolution by pervasive hydrothermal fluids of pre-existing carbonates and phosphates associated with organic matter. These iron-oxide spherules have been attributed to a later hydrothermal event at approximately 2.52 Ga (Orberger et al., 2012).

2.2. Sample preparation, and isotope and element analyses

The BIF sample was first cut into several columns, with cross-sections typically of $0.5 \times 1\ \text{cm}^2$, assigned to nitrogen and argon isotope analyses, iron isotope analyses, and major and trace element analyses. The band samples, typically weighing 0.3–1 g, were obtained by slicing the columns band-to-band. Only the summaries for these analyses, performed using various instruments at various laboratories, are described in the main text, whereas details in procedures and performances for these analyses are described in the supplementary part or in the references.

Volatile elements were extracted from homogenized powder of rock samples, using a stepwise combustion method to separate modern contamination from N indigenous to the sample. Isotopic compositions of $^{14,15}\text{N}$ and $^{36,40}\text{Ar}$, and abundances of C

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