



Extremely low oxygen concentration in mid-Proterozoic shallow seawaters



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ABSTRACT

The mid-Proterozoic (1.8–0.8 Ga) witnessed the first appearance but unusually low diversification of eukaryotes. The stagnant biotic evolution during this billion-year-long period (commonly referred to as the “Boring Billion”) was arguably ascribed to low oxygen levels in atmosphere and ocean. However, evidence supporting low oxygen in shallow-marine environments where early eukaryotes first evolved is generally lacking or insufficient. Here we report Rare Earth Element and yttrium (REE + Y) data, particularly cerium (Ce) anomalies, from a suite of mid-Proterozoic sedimentary rocks of the North China platform. The new data from North China, in combination with available Ce anomaly data from other Proterozoic successions, demonstrate that during mid-Proterozoic, negative Ce anomalies did not occur until ~1.54 Ga and after ~1.54 Ga, only episodic negative Ce anomalies were present in shallow-water carbonates. Trace element enrichments (U_{EF} , V_{EF} , and Mo_{EF}) remained at the average continental crust level before ~1.54 Ga but showed a sudden increase at ~1.54 Ga. The data suggest that oxygen concentration in shallow-marine environments of the mid-Proterozoic ocean was extremely low, probably <0.2 μ M prior to ~1.54 Ga (based on minimal oxygen concentration requirement for Ce(III) oxidation) and fluctuating around 0.2 μ M afterwards. The low oxygen concentration (~0.2 μ M) in shallow waters of the mid-Proterozoic ocean accounts for only ~0.1% of the modern surface ocean oxygen level (~280 μ M) and may help explain the evolutionary stasis of eukaryotes during the mid-Proterozoic.

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1. Introduction

Increasing evidence in the last decade has shown that the oxygenation of Earth’s surface environments occurred at the beginning and end of the Proterozoic (Och and Shields-Zhou, 2012; Lyons et al., 2014). During the Great Oxidation Event (GOE; Holland, 2002) around 2.4 Ga (e.g., Bekker et al., 2004), atmospheric oxygen may have increased from <0.001% present atmospheric level (PAL) in Archean to >0.1% PAL (e.g., Farquhar et al., 2000; Pavlov and Kasting, 2002; Holland, 2006) in Paleoproterozoic, resulting in stratified oceans with possibly oxic surface water but ferruginous or euxinic deep-water (e.g., Canfield, 1998; Planavsky et al., 2011; Reinhard et al., 2013; Lyons et al., 2014). During the Neoproterozoic Oxygenation Event (NOE; Och and Shields-Zhou, 2012) between 800 and

542 Ma (e.g., Fike et al., 2006; Canfield et al., 2007; Scott et al., 2008; Sahoo et al., 2012; Kendall et al., 2015), atmospheric oxygen level further increased (e.g., Canfield and Teske, 1996; Holland, 2006; Lyons et al., 2014), leading to well-oxygenated shallow-waters and variably oxygenated deep-waters in the ocean (e.g., Canfield et al., 2008; Johnston et al., 2013; Kurzweil et al., 2015; Sperling et al., 2015).

The mid-Proterozoic (1.8–0.8 Ga) between GOE and NOE is an intriguing period in Earth history that witnessed the first appearance but unusually low diversity of primitive eukaryotes (Han and Runnegar, 1992; Rasmussen et al., 2008). Although some multicellular eukaryotes appeared during the late Mesoproterozoic (Woods et al., 1998; Butterfield, 2000, 2001), the evolution of eukaryotic organisms was apparently stagnant (Knoll et al., 1995, 2006; Javaux et al., 2001; Lamb et al., 2009) and their abundance was very low (Brocks et al., 2005; Blumenberg et al., 2012; Reinhard et al., 2013). This billion-year-long period of evolutionary stasis, commonly referred to as the “Boring Billion”, was thought to be related to low oxygen levels in atmosphere and ocean (e.g., Anbar and Knoll, 2002; Canfield, 2005; Knoll et al., 2006; Kump, 2008;

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Reinhard et al., 2013; Smith and Harper, 2013; Luo et al., 2014; Planavsky et al., 2014; Gilleaudeau and Kah, 2015), although some have argued that the evolution of life is intrinsically controlled and independent of surface oxygenation (e.g., Butterfield, 2009; Erwin et al., 2011; Sperling et al., 2013; Mills et al., 2014).

The redox landscape of the mid-Proterozoic ocean, however, remains debated. Earlier studies suggested that the ocean was oxygenated after ~1.8 Ga, coincident with the disappearance of banded iron formation (BIF) (e.g., Holland, 2006). In another model, the mid-Proterozoic ocean was envisioned to be stratified and dominated by H₂S (euxinic conditions) in the deep ocean (Canfield, 1998). More recent studies demonstrated widespread iron-rich (ferruginous) conditions in the mid-Proterozoic ocean (Planavsky et al., 2011; Reinhard et al., 2013; Lyons et al., 2014). Estimation of mid-Proterozoic atmospheric O₂ level also varies significantly. Traditional pO₂ estimates of 1–40% PAL (e.g., Holland, 2002, 2006) were well above the oxygen demands of eukaryotes. More recent studies suggested that pO₂ may have declined significantly (to <0.1% PAL) after the GOE and remained at this level for more than a billion years (Bekker and Holland, 2012; Lyons et al., 2014; Planavsky et al., 2014). This extremely low (<0.1% PAL) pO₂ estimate raises questions about whether there was sufficient O₂ to oxidize surface ocean seawater and promote the diversification of eukaryotes. Answering these questions requires understanding the redox conditions of shallow-marine environments in the mid-Proterozoic ocean.

Rare Earth Elements and yttrium (REE+Y) data, particularly cerium (Ce) anomalies (expressed as Ce/Ce* = [Ce_{(SN)3}]/(Pr_{(SN)3}²/Nd_{(SN)3}); Lawrence et al., 2006), provide a means of documenting shallow-marine redox conditions (e.g., Planavsky et al., 2010; Ling et al., 2013). Unlike other REEs that typically exist in a trivalent oxidation state, Ce is unique in that it can also exist in tetravalent oxidation state, depending on environmental redox conditions (de Baar et al., 1988; German and Elderfield, 1989; Sholkovitz and Schneider, 1991). In modern oxygenated seawater, Ce(III) is rapidly oxidized to Ce(IV) through mediation of manganese oxide and/or bacteria (Byrne and Sholkovitz, 1996; Tachikawa et al., 1999; Tanaka et al., 2010). Ce(IV) is adsorbed by Fe–Mn nodules and crusts in oxygenated deep ocean (Elderfield et al., 1981; Bau et al., 1996), resulting in negative Ce anomalies in seawater and positive Ce anomalies in Fe–Mn oxides (Bau et al., 1996). Although Ce concentrations in shallow seawaters are highly variable (Kamber and Webb, 2001), highest Ce/Ce* values of the surface waters in modern oceans or seas are all less than 0.55 (Shimizu et al., 1994; Bertram and Elderfield, 1993; German et al., 1995; Nozaki et al., 1999; Slack et al., 2007). In anoxic oceans such as those in Archean and Paleoproterozoic, the lack of Ce(III) oxidation and absence of deep-ocean Fe–Mn oxides lead to high seawater Ce concentration. Therefore, no negative Ce anomaly is expected and Ce/Ce* values may be close to or higher than 1.0 (Kamber and Webb, 2001; Van Kranendonk et al., 2003). Although some negative Ce anomalies were reported from Archean sedimentary rocks, their veracity and depositional origin have been questioned (e.g., Planavsky et al., 2010). Intermediately oxygenated ocean may have Ce/Ce* values between 0.55 and 1 (e.g., Ling et al., 2013). Thus, secular Ce/Ce* record tracks first-order ocean redox changes in Earth history.

Within the envelope of secular Ce/Ce* record, Ce anomalies can also record local redox conditions. In redox-stratified environments (basins), reductive dissolution of settling Fe–Mn oxides below Fe–Mn redox boundaries lead to Ce(III) enrichments and higher Ce/Ce* values in deep waters below the chemocline (German et al., 1991; Byrne and Sholkovitz, 1996; Slack et al., 2007). In this case, lower Ce/Ce* values in shallow water environments at or above the chemocline are expected. When the entire water column has low level of dissolved oxygen (without highly

oxygenated shallow waters), Ce redox-cycle may stop and differential Ce/Ce* values are not expected between shallow- and deep-water environments.

In this paper, we use Ce anomaly and REE patterns, along with redox-sensitive trace element enrichments (U, V, and Mo), to evaluate the redox evolution of shallow waters on the North China platform during the period of ca. 1.65–1.47 Ga. We also construct the secular Ce/Ce* record with available data from other Proterozoic successions. To date, the shallow-water redox state of the mid-Proterozoic ocean has not been sufficiently investigated, except for the recent study of Cr isotopes (Planavsky et al., 2014) and iodine concentrations of Archean and early Paleoproterozoic carbonates (Hardisty et al., 2014). We aim at testing the hypothesis that the low oxygen level in surface ocean during mid-Proterozoic has prevented the diversification of early eukaryotes.

2. Geological setting

2.1. Regional geology

Associated with the break-up of supercontinent Columbia or Nuna (Zhao et al., 2003, 2004, 2011; Zhang et al., 2012) to the assembly of Rodinia (Li et al., 2008), a thick (~9000 m) sedimentary succession was deposited on the North China platform during the Proterozoic. This succession is commonly subdivided into three groups (Fig. 1) including the Changcheng Group (1800–1600 Ma, Pt₁), Jixian Group (1600–1400 Ma, Pt₂), and the Qingbaikou Group (1000–800 Ma, Pt₃), with a significant hiatus of ~400 Ma between the Jixian and Qingbaikou groups (Gao et al., 2009; Su et al., 2010; Li et al., 2013; Su, 2014).

The Changcheng Group is dominated by siliciclastic rocks that were deposited on the Archean-early Paleoproterozoic crystalline basement. This group, in stratigraphic order, includes the Changzhougou, Chuanlinggou, Tuanshanzi and Dahongyu formations. Alluvial to fluvial conglomerates and sandstones constitute the basal part of the group, representing deposition associated with the initial regional extension (Lu et al., 2002, 2008). Ironstones and black shales of the Chuanlinggou Formation probably record the initial breakup of the Columbia supercontinent and a rapid basement subsidence (Zhao et al., 2011; Tang et al., 2015). The Jixian Group includes the Gaoyuzhuang, Yangzhuang, Wumishan, Hongshuizhuang and Tieling formations and is dominated by carbonate rocks including laminated and stromatolitic dolostone, limestone, argillaceous dolostone and chert bands. The thick carbonate succession was likely deposited from an epicontinental sea. Above the Jixian Group, a regional unconformity is observed across the entire North China platform (Qiao et al., 2007; Su et al., 2010). The overlying Xiamaling Formation consists mainly of organic-rich black shales that were likely deposited from low-energy environments of an intra-shelf basin (Qiao et al., 2007; Luo et al., 2014). Unconformably overlying the Xiamaling Formation is siliciclastic and carbonate rocks of the Changlongshan and Jing'eryu formations of the Qingbaikou Group. A significant uplift after the Jing'eryu Formation exposed the Proterozoic strata until the middle Cambrian (Zhou et al., 2006), which is commonly thought to have resulted from the formation of supercontinent Rodinia (Wang et al., 2000; Lu et al., 2008).

The mid-Proterozoic strata are well preserved on the North China platform, with low metamorphism commonly below prehnite–pumpellyite phase (Li et al., 2003; Chu et al., 2007). The low metamorphism of rocks warrants that REE+Y signatures may record their primary signals (Bau and Möller, 1993) and can be used for paleoenvironmental interpretation (e.g., Planavsky et al., 2010; Ling et al., 2013).

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