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Uranium in iron formations and the rise of atmospheric oxygen

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ABSTRACT

The concept of the Great Oxidation Event (GOE), during which atmospheric oxygen rose precipitously and perhaps to near-modern levels around 2.4–2.1 billion years ago (Ga), has become entrenched in our views on secular atmospheric evolution. Multiple proxies confirm a permanent shift towards more oxygenated conditions at some time near the Archean–Proterozoic boundary. However, it remains unclear precisely when this transition occurred, due in part to the likely temporal variability in those early levels and different sensitivities of the proxies utilized to track atmospheric oxygen partial pressures. Here, we provide a new look at the timing and magnitude of early atmospheric oxygenation through the record of uranium (U) concentrations in iron formations (IF). Just as IF are important archives of the redox state of seawater, concentrations of redox-sensitive U in IF are faithful proxies for oxidative continental weathering and associated delivery of dissolved U to seawater. Our dataset suggests that there was an increase in U redox cycling and transport at ca. 2.47 Ga, just before the permanent loss of mass-independent sedimentary sulfur isotope anomalies traditionally used to define the onset of the GOE. Further, there is significant temporal variability in the IF U record that we propose reflects dynamic Precambrian redox conditions. We provide additional support for earlier suggestions that the GOE was a protracted event marked by vacillating oxygen levels.

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1. Introduction

From its inception, the concept of an initial, irreversible rise of atmospheric oxygen beyond some threshold value, the so-called ‘Great Oxidation Event’ (GOE; Holland, 2002), has been difficult to define in terms of its timing, duration, and magnitude. This remains so despite decades of work, a relationship likely mirroring the complexity of the event. Dick Holland's work has underpinned our basic understanding of the GOE, which was aptly named by him for the dramatic geochemical changes he recognized to be associated with this ‘event.’ Without question, Dick Holland fundamentally changed our knowledge of oxygenation trends through time; we honor that contribution by continuing to refine those trends. Presently, numerous paleoredox proxies suggest that the first transition from an anoxic to an oxic atmosphere, and the first widespread oxidative weathering of the continents, occurred transiently right at the Archean–Proterozoic boundary at 2.5 Ga (e.g., Anbar et al., 2007; Kump, 2008; Reinhard et al., 2009; Duan et al., 2010; Kendall et al., 2010), and then as a more prominent process by around 2.48 Ga (Konhauser et al., 2011). Atmospheric oxygen concentrations are thought to have reached 1 to 10% of present atmospheric level (PAL), and perhaps much higher, by ca. 2.3–2.1 Ga (Holland, 1994; Bekker et al., 2004; Kump, 2008; Bekker and Holland, 2012). Despite these basic constraints, the exact timing and temporal fabric of the GOE remain poorly constrained. This work strives to better define the first rise in atmospheric O_2 related to the GOE.

Typically, the GOE has been defined as a single event (cf. Holland, 2002) starting with the loss of mass-independent sulfur isotope fractionation (S-MIF) from the sedimentary record between ca. 2.45 and 2.32 Ga (Farquhar et al., 2000; Bekker et al., 2004; Guo et al., 2009). The S-MIF signal is generally attributed to low atmospheric oxygen levels, $<10^{-5}$ PAL according to atmospheric models (Pavlov and Kasting, 2002), or, alternatively, to methane mixing ratios above 3×10^{-5} bars (Zahnle et al., 2006). The GOE might instead be viewed as a more protracted event marked by rising and falling oxygen levels rather than a secular change marked by the loss of S-MIF. We might, therefore, think of the GOE as an interval of time rather than an event, which included earlier stages of continental oxidation tied to the initial rise of atmospheric oxygen before the ultimate loss of S-MIF (e.g., Anbar et al., 2007; Konhauser et al., 2011; Reinhard et al., 2013a).

Given the dynamic geochemical backdrop of the GOE, each paleoredox proxy may offer unique minimum or maximum constraints on atmospheric oxygen level and the redox state of the oceans because of differing redox sensitivities. Accordingly, redox-sensitive elements

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could have varied in their responses to the rise in atmospheric oxygen. For example, significant U enrichment in shales first occurs in the Timeball Hill and Rooihoogte formations of South Africa (Partin et al., 2013), the units immediately above the Duitschland Formation (Bekker et al., 2004) where the loss of S-MIF is recorded (Bekker et al., 2004; Guo et al., 2009). There is also a shift in the extent of iron isotope variability in sedimentary rocks interpreted to reflect a change in oceanic redox state at around 2.35 Ga (Rouxel et al., 2005; Planavsky et al., 2012b). However, there are also notable stratigraphic Mo and Re enrichments in the much older 2.5 Ga Mount McRae Shale of Western Australia (Anbar et al., 2007), in addition to changes in sedimentary sulfur (Kaufman et al., 2007), nitrogen (Garvin et al., 2009), and molybdenum isotope compositions (Duan et al., 2010) that point to at least mild oxidative weathering conditions during the deposition of this unit, which may have resulted in H_2S-bearing (euxinic) water column conditions with enhanced primary productivity (Reinhard et al., 2009; Raiswell et al., 2011). Similar geochemical observations and conclusions have been made for shallow-water facies of the coeval Klein Naute Formation in the Griqualand West basin of South Africa (Kendall et al., 2010). Furthermore, the U isotope composition of the Mount McRae Shale has been interpreted to be consistent with U mobility at that time, implying oxidative weathering and delivery of U to the oceans (Kendall et al., 2013–in this issue). Partial oxygenation, that is enough oxygen to permit some oxidative continental weathering, could have occurred before the rise of atmospheric oxygen as defined by the loss of S-MIF, or alternatively, continuation of the S-MIF signal through sedimentary recycling after oxygen levels rose above 10^{-5} PAL (Reinhard et al., 2013a). In this scenario, the S-MIF signal could be derived from oxidative weathering of older (Archean) sedimentary sulfides after the 10^{-5} PAL threshold was exceeded (Reinhard et al., 2013a). With this in mind, it is possible that atmospheric oxygen levels were above 10^{-5} PAL for periods from 2.5 to 2.32 Ga despite the persistence of the sedimentary S-MIF signal throughout that period. The coexistence of the S-MIF signal and at least partial oxygenation is evidenced by extensive Cr mobilization at ca. 2.48 Ga (Konhauser et al., 2011), transient terrestrial sulfide oxidation at ca. 2.5 Ga (e.g., Anbar et al., 2007; Reinhard et al., 2009; Duan et al., 2010; Kendall et al., 2010), and perhaps even earlier (Wille et al., 2007; Voegelin et al., 2010) based on Mo isotope systematics in carbonate and shale, and Re abundances in shale (Kendall et al., 2010).

Further complicating the story of Paleoproterozoic redox evolution, several lines of evidence point towards a drop in post-GOE atmospheric oxygen level at Earth's surface. Specifically, the U record of anoxic shales suggests that atmospheric oxygen levels declined around 2.05 Ga (Partin et al., 2013). The sulfate evaporite and S isotope records also indicate a fall in atmospheric–oceanic oxygen levels between 2.1 and 2.0 Ga (Schröder et al., 2008; Bekker and Holland, 2012; Planavsky et al., 2012a).

Despite the emerging view of post-GOE atmospheric oxygen fluctuations, uncertainties remain. We aim to clarify the timing and structure of Earth's first oxygenation by exploring the redox-sensitive nature of U in seawater and its history as recorded by U enrichments in iron formations (IF), which are iron- and silica-rich chemical sediments deposited throughout much of the Precambrian. Because these sediments precipitated directly from seawater, their chemical composition has been widely used as a proxy for ancient seawater composition. For instance, the IF record has been used to decipher first-order trends in the concentrations of Ni and P in Precambrian seawater (Bjerrum and Canfield, 2002; Konhauser et al., 2009; Planavsky et al., 2010). The IF record is particularly well-suited to recording variations in the U content of seawater as the geochemical cycles of U and Fe are closely linked, with the U(VI)/U(IV) and Fe(III)/Fe(II) redox couples being similar in reduction potential (e.g., Barnes and Cochran, 1993; Bruno et al., 1995). Most U in IF is likely to have been derived directly from seawater by adsorption and co-precipitation processes (see 4. Discussion); it is this aspect of IF that confers their ability to track the content of soluble U(VI) in seawater. Utilizing the framework established by the U in shales record (Partin et al., 2013), which provides a high temporal resolution record of the changes in U seawater chemistry and oxidative continental weathering, the IF record provides an opportunity to further explore changes in the U record related to the GOE by bridging the two proxies. While the U concentration of both IF and shales is limited by concentration of U in contemporaneous seawater, each reflects two separate, coexisting sinks that, combined, provide a more complete picture of marine U cycling through time. Additionally, both lithologies are nonuniformly preserved in the geologic record, and therefore, one can be used to fill temporal gaps in the other, e.g., the IF record fills a critical gap in the available 2.45 to 2.32 Ga shale record. Although the two lithologies have notably different depositional styles and temporal distribution, the IF and shale records show remarkably similar trace element histories, reinforcing the view that both lithologies archive changes in seawater composition.

2. Methods

The dataset of iron formation and iron-rich chemical sediments presented here is comprised of both new analyses and a comprehensive literature compilation (see Table 1 for a list of IF units with U data available and Supplementary data Table S1 for sample values). Samples with a high degree of weathering, alteration, metamorphic or diagenetic overprinting were avoided. Specifically, the data were filtered for metamorphic grade (upper greenschist facies or below, as a baseline to maintain a comparable mineralogy between samples), sulfur content (<1%), iron content (5–60%), and compatible mineralogy—in other words, limited to Fe- and Si-rich chemical sediments and excluding sulfides and carbonates. The filtered dataset includes 1315 U concentration values, 591 of which are derived from analysis of bulk powders after digestion, and 724 from laser ablation of polished thin sections; an additional 148 values are included in Table S1 that pass all filter criteria except metamorphic grade (see Fig. 1A). For new data presented here, drill core and hand samples were cleaned and then powdered in an agate mill or processed into thin sections (see Konhauser et al., 2011 for further details). Data were acquired at the University of Alberta using a PerkinElmer Elan6000 Quadrupole Inductively Coupled Plasma Mass Spectrometer (Q-ICP-MS) in solution mode (after heated HF–HNO_3 digestion) or by in-situ laser ablation with a New Wave Research UP-213 laser. Precision was monitored by repeated analyses of international standards BE-N Basalt (CRPG Nancy) in solution mode and NIST glasses SRM 610 and 612 by laser ablation. At the Woods Hole Oceanographic Institution (WHOI) and IFREMER (French Research Institute for Exploitation of the Sea, Brest), samples were sequentially digested in heated HF + HNO_3, *aqua regia*, and 6 N HCl with trace H_2O_2. Data were acquired in solution mode using a ThermoElectron Inc. Element 2 double focusing magnetic sector field ICP-MS and were normalized against repeated analyses of international standards IF-G (CRPG Nancy) and BHVO-1 (USGS). The *in situ* grain-by-grain laser-ablation data (from hematite and magnetite) should give more insight into the U concentrations associated with the iron oxide mineral phase itself, assumed to reflect the vestige of precursor ferric oxyhydroxide minerals (such as ferrihydrite), as opposed to the U concentration of the bulk sample; the data obtained with these two analytical methods provides a useful comparison of mineral-specific U concentrations relative to conventional bulk analysis U concentrations. Precision in U concentration analyses is estimated to be better than 10% for all data. Radioactive decay causes some U and Th to be lost over geological time; U and Th concentrations were corrected for radioactive decay with respect to their host sediment age by assuming that the majority of U is ^{238}U and the majority of Th is ^{232}Th as follows (e.g., Faure, 1986): $[U]_{corrected} = [U]_{measured} \times e^{0.155125 \times Age\ (Ga)}$ and $[Th]_{corrected} = [Th]_{measured} \times e^{0.049475 \times Age\ (Ga)}$. For the oldest samples, corrected U and Th concentrations were 1.8× and 1.2× measured concentrations, respectively.

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