



Enhanced organic carbon burial in large Proterozoic lakes: Implications for atmospheric oxygenation

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ABSTRACT

The burial of organic carbon in sedimentary systems has been a fundamental part of the carbon cycle throughout the geological record, and was instrumental in major oxygenations of the atmosphere in the early Palaeoproterozoic and Neoproterozoic. While much focus has been placed on the burial of carbon in Precambrian marine carbonate and organic carbon-rich rocks deposited around the time of these major oxygenations, such deposits yield little information on the evolution of the atmosphere in the significant time between. There is, however, growing evidence from terrestrially deposited sediments to suggest the surface environment may have been at least intermittently well-oxygenated from the late Mesoproterozoic. Hence Proterozoic sediments deposited in terrestrial near-surface environments are useful targets for the study of atmospheric evolution during a time which is hitherto poorly understood.

Thus far, little attention has been paid to the contribution of large lakes and intercontinental basins to the global burial of organic carbon, and thus the progressive oxygenation of the atmosphere, especially given that the highest rates of organic carbon burial in modern aquatic environments occur in lacustrine settings, in stark contrast to the low rates observed in the contemporary marine realm. Here, we report high burial rates of organic carbon in large lacustrine systems of late Mesoproterozoic to early Neoproterozoic age, which are comparable with modern lacustrine systems, and significantly higher than modern and ancient marine deposits. These data emphasise the significance of lakes as a global repository for organic carbon, and imply Proterozoic lakes were at least as efficient, and perhaps as important, as modern lakes in the global burial of organic carbon. Such findings suggest large Proterozoic lakes and epicontinental basins played a crucial role in the progressive oxygenation of the atmosphere before the major Neoproterozoic oxygenation.

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1. Introduction

1.1. Carbon burial

The burial of organic carbon within sediments is a fundamental aspect of the carbon cycle, and has consequences for the carbon isotopic record (Schidlowski, 1991), the isotopic composition of the oceans (Tosca et al., 2010), the composition of the atmosphere (Lenton and Watson, 2004; Doney and Schimel, 2007), the budget of other elements including phosphorus and sulphur (Bernier

and Raiswell, 1983; Schuffert et al., 1998; Gill et al., 2007) and the deep biosphere (Wellsbury et al., 1997). Modelling of organic carbon burial is underpinned by data for specific examples in present day environments and in the geological record (Müller and Suess, 1979; Dean and Gorham, 1998; Hayes et al., 1999; Burdige, 2007). However, such data are sparse for deep geological time (Campbell and Allen, 2008). Large positive carbon isotopic excursions in marine sediments deposited ~2.3 Ga and between ~830 and 490 Ma are attributed to increases in the global burial of organic carbon (Shields and Veizer, 2002; Halverson et al., 2005; Holland, 2006; Shields-Zhou and Och, 2011; Och and Shields-Zhou, 2012) during times which, crucially, are thought to have witnessed the first two large atmospheric oxygenations (e.g. Holland, 2006; Kah and Bartley, 2011), thereby highlighting the importance of organic carbon burial to the oxygenation of the atmosphere. However, these large isotopic excursions are recorded in marine carbonate rocks.

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Non-marine depocentres in terrestrial intra-cratonic basins may have hosted conditions that were favourable for organic carbon burial at times other than those reflected in the marine sedimentary rock record. Such basins are geochemically independent of their marine counterparts, and, crucially, are generally subject to higher sedimentation rates. This in turn raises the possibility that, in the time between the two large positive carbon isotopic excursions, the rate of organic carbon burial may have been higher in some terrestrial basins than in coeval marine basins, thus facilitating the evasion of oxygen to the atmosphere.

1.2. Overview of pre-Phanerozoic atmospheric oxygenation

The timing of the oxygenation of the Earth's atmosphere is a subject of intense interest among Precambrian workers, and is not fully agreed upon. That said, two distinct rises in free atmospheric oxygen are thought to have occurred prior to the Phanerozoic: the first during the Archaean–Proterozoic Transition from 2.6 to 2.4 Ga, and in the second in the Neoproterozoic ~0.85 Ga. The oxidation history between these distinct rises, during the Mesoproterozoic, however, is far less well constrained.

The first major rise in atmospheric oxygen, widely referred to as the 'Great Oxidation Event' (GOE; Holland, 2002, 2006), was postulated to represent the first irreversible introduction of biogenic oxygen to the atmosphere at around 2.3 Ga, evident largely from high ^{13}C carbon isotope signatures in marine carbonates (Karhu and Holland, 1996; Shields and Veizer, 2002; Halverson et al., 2005), a transition of mass-independent fractionation (MIF) of sulphur to mass-dependent fractionation (MDF) of sulphur (Pavlov and Kasting, 2002; Guo et al., 2009), and the appearance of terrigenous sedimentary red beds coincidental with the disappearance of redox-sensitive detrital minerals such as uraninite and pyrite in terrigenous sediments (Holland, 1984, 1999). The transition from MIF to MDF of sulphur in sediments ~2.3 Ga suggests the Earth's atmospheric free oxygen rose to at least 0.001% of present atmospheric level (PAL; Pavlov and Kasting, 2002), before which it is thought to have remained below 0.001% PAL (Fig. 1). Other estimates for the atmospheric oxygen concentration immediately after ~2.30 Ga from the rate of terrestrial oxidative weathering and from the evidence for anoxic oceans suggests an atmospheric oxygen concentration of 1–10% PAL (Kump, 2008). Recent studies suggest an extreme increase in atmospheric oxygen at ~2.3 Ga, possibly to levels similar to modern levels, followed by a large drop a few hundred million years later (Fig. 1; Lyons et al., 2014). There is complex and contradicting evidence both for atmospheric anoxia before the GOE and for the sustained presence of atmospheric oxygen immediately following the GOE. Thus constraining the timing of the introduction of free oxygen to the atmosphere is, at present, problematic. Geochemical data from redox-sensitive metals in sedimentary rocks suggest there may have been at least localised oxidised surface environments as early as ~3 Ga (Wille et al., 2007; Frei et al., 2009; Crowe et al., 2013). Likewise, there is evidence for a decline in the concentration of atmospheric oxygen following the GOE (Frei et al., 2009). The combined geochemical evidence for atmospheric oxygen flux either side of the postulated GOE suggests that the initial oxidation of the atmosphere was episodic, resulting in numerous transient periods of surface oxidation from ~3 Ga until well into the Proterozoic. Thus the first atmospheric oxidation was a long-lived transition, rather than an event.

The second major step in atmospheric oxygenation is traditionally thought to have occurred from ~0.85 Ga (Canfield, 2005; Holland, 2006), during which oxygen accumulated to levels high enough to allow the evolution of complex multicellular life such as metazoa (Knoll and Carroll, 1999) from ~635 Ma (Hofmann et al., 1990; Love et al., 2009). Geochemical evidence for this step in atmospheric oxygenation include globally high ^{13}C signatures in

marine carbonates deposited between ~830 and 490 Ma (Shields and Veizer, 2002; Halverson et al., 2005) which suggest a rapid increase in the burial of organic carbon and the concomitant release of oxygen. Large fractionations in sulphur isotopes from ~580 Ma onwards (Canfield and Teske, 1996; Canfield, 1998) which indicate an increase in the oxidation of sulphide to sulphate and an increase in the oxidative part of the sulphur cycle, are strong geochemical indicators for a large increase in atmospheric oxygen during the Neoproterozoic. Recent evidence for an increase in atmospheric oxygen during the Neoproterozoic is derived from redox-sensitive trace metals. An increase in the concentration of metals such as Mo, V and U in marine black shales (Scott et al., 2008; Campbell and Allen, 2008; Shields-Zhou and Och, 2011; Och and Shields-Zhou, 2012), and positive fractionations of chromium isotopes in Fe-rich cherts (Frei et al., 2009) provide strong evidence for oxidative weathering of terrigenous metals during the Neoproterozoic. Estimates for the atmospheric oxygen concentration during the Neoproterozoic oxygenation range from 10% to 20% PAL (e.g. Canfield, 2005; Holland, 2006; Lyons et al., 2014), though this concentration may have fluctuated significantly (Laakso and Schrag, 2014).

While much work has been dedicated to the GOE and Neoproterozoic oxygenation, relatively little emphasis has been placed on the significant time between. Traditionally this period has been thought to have represented a time of atmospheric evolutionary stasis (Buick et al., 1995; Brasier and Lindsay, 1998; Holland, 2006) with little change in the redox state of the atmosphere. This notion was largely based upon the marine carbon isotope record showing little change (Buick et al., 1995). However, there is a growing body of evidence to suggest that towards the end of the Mesoproterozoic, between ~1.3 and 1.2 Ga, there were at least localised oxidised surface environments. During this period the marine sulphate concentrations rose from 1.5–2.4 mM to 2.7–4.5 mM (Hurtgen et al., 2002; Campbell and Allen, 2008), suggesting an increase in the rate of oxidative weathering of terrestrial sulphides. This is also reflected in the first appearance of widespread marine gypsum deposits (Kah et al., 2001). The appearance of microbial disproportionation of sulphur in marine sediments between ~1.3 and 1.2 Ga (Johnston et al., 2005) similarly suggests enhanced oxygenation of the marine environment, thus oxygenation of the atmosphere. A limitation on the geochemical proxies for atmospheric oxygenation throughout the Mesoproterozoic derives from the redox state of the oceans. There is broad agreement that the euxinic conditions were widespread throughout much of the Mesoproterozoic (e.g. Canfield, 1998; Lyons et al., 2009), though perhaps not ubiquitous, as ferruginous conditions may also have existed at least locally (Planavsky et al., 2011; Kah and Bartley, 2011). Such sulphidic conditions in the oceans may have sequestered redox-sensitive and bioessential trace metals such as molybdenum (Anbar and Knoll, 2002; Scott et al., 2008; Och and Shields-Zhou, 2012; Gilleaudeau and Kah, 2013), the scarcity of which may have limited the evolution of microbial life in the marine environment (Parnell et al., 2012), and thus limited the biogenic generation of oxygen through photosynthesis, and organic carbon burial. Bioessential metals, geochemical ingredients critical for biological processes, could have been less chemically sequestered in continental aquatic environments, thus increasing the potential for their bioavailability. Furthermore, their delivery to the surface environment could have been enhanced due to oxidative weathering of increasingly metalliferous anorogenic granites within supercontinents from ~1.9 Ga (Parnell et al., 2012), the formation of which has been linked with coeval distinct increases in atmospheric oxygen (Campbell and Allen, 2008). Enhanced metal delivery to surface environments via oxidative weathering of terrestrial sulphides can be constrained by the increased delivery of sulphate to the oceans from ~1.7 to 1.6 Ga (Grotzinger and Kasting,

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