



Ocean redox structure across the Late Neoproterozoic Oxygenation Event: A nitrogen isotope perspective



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ABSTRACT

The end of the Neoproterozoic Era (1000 to 541 Ma) is widely believed to have seen the transition from a dominantly anoxic to an oxygenated deep ocean. This purported redox transition appears to be closely linked temporally with metazoan radiation and extraordinary perturbations to the global carbon cycle. However, the geochemical record of this transition is not straightforward, and individual data sets have been variably interpreted to indicate full oxygenation by the early Ediacaran Period (635 to 541 Ma) and deep ocean anoxia persevering as late as the early Cambrian. Because any change in marine redox structure would have profoundly impacted nitrogen nutrient cycling in the global ocean, the N isotope signature of sedimentary rocks ($\delta^{15}\text{N}_{\text{sed}}$) should reflect the Neoproterozoic deep-ocean redox transition. We present new N isotope data from Amazonia, northwest Canada, northeast Svalbard, and South China that span the Cryogenian glaciations (~750 to 580 Ma). These and previously published data reveal a N-isotope distribution that closely resembles modern marine sediments, with a mode in $\delta^{15}\text{N}$ close to +4‰ and range from -4 to +11‰. No apparent change is seen between the Cryogenian and Ediacaran. Data from earlier Proterozoic samples show a similar distribution, but shifted slightly towards more negative $\delta^{15}\text{N}$ values and with a wider range. The most parsimonious explanation for the similarity of these N-isotope distribution is that as in the modern ocean, nitrate (and hence O_2) was stable in most of the middle-late Neoproterozoic ocean, and possibly much of Proterozoic Eon. However, nitrate would likely have been depleted in partially restricted basins and oxygen minimum zones (OMZs), which may have been more widespread than in the modern ocean.

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1. Introduction

It is widely argued that the appearance of animals in the geological record is linked to a major step in the oxygenation of Earth's surface environment, known as the Neoproterozoic Oxygenation Event (NOE) (Canfield et al., 2007, 2008; Och and Shields-Zhou, 2012; Scott et al., 2008). The NOE is more difficult to document in the geological record than its early Paleoproterozoic counterpart, the Great Oxidation Event (GOE), whose onset is well established based on the sedimentary record of redox-

sensitive detrital minerals and mass independent fractionation of sulfur isotopes (e.g. Farquhar et al., 2000; Lyons et al., 2014). Although the NOE has not been formally defined, one criterion is the timing of ventilation of the deep ocean with sufficient dissolved oxygen to accommodate the earliest animals (Och and Shields-Zhou, 2012). Whereas the cause-and-effect relationship between oxygenation and animal evolution and the oxygen threshold required for early animals are still debated (e.g., Butterfield, 2009; Sperling et al., 2013), eukaryotic diversification and the oxygenation of the environment must have been closely coupled through a complex network of biogeochemical feedbacks linked through the origin, export, decay and burial of organic matter (Butterfield, 2009; Close et al., 2011; Logan et al., 1995; Pawlowska et al., 2013).

Iron speciation data from Neoproterozoic mudstones suggest a transition from dominantly anoxic and ferruginous deep oceans

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with prevalent sulfidic (euxinic) conditions in continental margin settings (Planavsky et al., 2011; Poulton and Canfield, 2011) to dominantly anoxic and ferruginous deep oceans with only occasional evidence for euxinic conditions in the early–middle Neoproterozoic (Canfield et al., 2008), followed by deep-ocean ventilation in the late Ediacaran (Canfield et al., 2007). However, these data only provide information about local depositional redox conditions and do not bear directly on the state of the deep ocean. Trace metal abundances (Mo, U, V, and Cr) in euxinic sediments, although sensitive to local conditions, are also strongly dependent on the volume of oxygenated water masses, hence they bear on global redox conditions in the oceans (Algeo and Rowe, 2012). A series of separate studies and compilations of trace metal data have been published recently. All show a dramatic increase in abundances in the Ediacaran first interpreted to record deep ocean ventilation (635–541 Ma; Scott et al., 2008; Och and Shields-Zhou, 2012; Sahoo et al., 2012; Partin et al., 2013), broadly consistent with iron speciation data. Yet, data from some individual basins variably indicate persistently anoxic (Li et al., 2010; Johnston et al., 2013) versus oxygenated (Johnston et al., 2012; Sansjofre et al., 2014) bottom-waters throughout most of the Ediacaran Period. Other data imply widespread oxygen-deficient conditions in the earliest Cambrian (e.g. Schroder and Grotzinger, 2007; Cremonese et al., 2013) despite the appearance of complexly burrowing bilaterians. An emerging model is that anoxic conditions on continental platforms was locally controlled (Kah and Bartley, 2011; Shen et al., 2011), the deep ocean being pervasively oxygenated, but with a low oxygen concentration preventing the stabilization of oxic conditions in platforms throughout the Ediacaran Period (Johnston et al., 2012, 2013). Recent modeling coupled with trace metal compilations further suggests that no more than 40% of the pre-NOE seafloor could have been anoxic and no more than 10% euxinic (Partin et al., 2013; Reinhard et al., 2013). These models and existing data thus allow for the possibility that much of the global ocean may have been oxygenated prior to the Ediacaran Period.

We have applied the nitrogen isotope system to the question of the NOE because the N-isotopic composition of marine sediments ($\delta^{15}\text{N}_{\text{sed}}$; where the standard delta notation and normalization to atmospheric N_2 is used; Mariotti et al., 1981) is strongly dependent on the nitrogen biogeochemical cycle in the water column, itself controlled by the ocean redox structure (Boyle et al., 2013; Canfield et al., 2010; Fennel et al., 2005; Quan and Falkowski, 2009). Although the present day nitrogen cycle is highly complex and remains poorly understood in detail (e.g. Lam and Kuypers, 2011; Sigman et al., 2009a), the current understanding of its basic features and of its isotopic imprint in the sedimentary record (Galbraith et al., 2008; Sigman et al., 2009b) have already motivated the application of $\delta^{15}\text{N}_{\text{sed}}$ to elucidate nitrogen biogeochemical cycling in past oceans. Nitrogen isotopes have been applied to the Phanerozoic (e.g. Algeo et al., 2008; Higgins et al., 2012; Junium and Arthur, 2007; Quan et al., 2008) and Precambrian records (Beaumont and Robert, 1999; Busigny et al., 2013; Garvin et al., 2009; Godfrey and Falkowski, 2009; Godfrey et al., 2013; Kump et al., 2011; Papineau et al., 2009, 2013; Stüeken, 2013; Thomazo et al., 2009, 2011) as a proxy for both ocean redox and nitrogen biogeochemistry. Here we add a new nitrogen isotope dataset from ~750 to 580 Ma strata from multiple cratons to a growing Neoproterozoic database of both bulk sedimentary ($\delta^{15}\text{N}_{\text{sed}}$) and kerogen ($\delta^{15}\text{N}_{\text{ker}}$) nitrogen isotope data (Cremonese et al., 2013; Kikumoto et al., 2014; Spangenberg et al., 2014) to test whether the NOE is recorded in nitrogen isotope signatures. These results show that the $\delta^{15}\text{N}_{\text{sed}}$ distributions for the Cryogenian and the Ediacaran resemble one another and that documented in the modern ocean.

1.1. Isotope biogeochemistry of the nitrogen oceanic cycle

Nitrogen-isotopic compositions of nitrate, particulate organic matter and surface sediments have been extensively studied to characterize the nitrogen cycle in present oceans and its transcription into the sedimentary record (Fig. 1a, e.g. Robinson et al., 2012; Somes et al., 2010; Tesdal et al., 2013; Thunell et al., 2004). Below we summarize the basic processes and pathways and associated isotope fractionation (expressed using the enrichment factor notation in ‰, $\epsilon_{a-b} \approx \delta^{15}\text{N}_a - \delta^{15}\text{N}_b$) in the oceanic nitrogen cycle.

The initial source of all bioavailable-N to the oceans is the fixation of atmospheric N_2 (N_2 -fixation) by aerobic or anaerobic autotrophs, such as cyanobacteria, which transform molecular N_2 into organic matter (via NH_4^+) with a small isotope fractionation ($\epsilon_{\text{org-N}_2} \sim -4$ to 0‰ ; e.g. Zerkle et al., 2008). Other primary photosynthetic organisms assimilate remineralized forms of nitrogen mostly as NO_3^- or NH_4^+ . The mineralization of organic matter produces NH_4^+ (ammonification) with no net isotope fractionation owing to its efficiency (Möbius, 2013). If NH_4^+ remains stable it can then be assimilated with an isotope effect that increases strongly with its availability ($\epsilon_{\text{org-NH}_4} \sim 0\text{‰}$ to -27‰) such that the organic matter can be markedly ^{15}N -depleted where NH_4^+ is abundant (Pennock et al., 1996). The NH_4^+ may also be oxidized. If this oxidation is not complete, the residual NH_4^+ will be enriched in ^{15}N , because the first oxidation step to nitrite (NO_2^-) involves a strong fractionation ($\epsilon_{\text{NO}_2-\text{NH}_4} \sim -41\text{‰}$ to -13‰ ; Casciotti et al., 2003; Mariotti et al., 1981; Santoro and Casciotti, 2011). In oxic environments, this oxidation is accomplished by nitrification (i.e. sequential oxidation to NO_2^- and NO_3^-) and goes to completion, unless ammonium is concurrently assimilated as a nutrient. Subsequent NO_3^- assimilation entails fractionation with $\epsilon_{\text{org-NO}_3}$ between 0 and -8‰ in NO_3^- -limited and NO_3^- -replete conditions, respectively (Pennock et al., 1996; Somes et al., 2010). Under dysoxic conditions (i.e. typically in OMZs or within redox transition zones), NO_3^- as well as available NH_4^+ and NO_2^- are converted into gaseous species (NO_2 and/or N_2) by an incompletely understood combination of metabolic pathways that include heterotrophic denitrification and anammox (anaerobic oxidation of NH_4^+ by NO_2^-), nitrification, nitrate reduction to NH_4^+ , chemolithotrophic sulfide-dependent denitrification, co-oxidation of methane and ammonium, and most probably other metabolisms yet to be identified (Lam et al., 2009; Lam and Kuypers, 2011; Lavik et al., 2009; Mandernack et al., 2009; Wenk et al., 2013). In present day OMZs where nitrate consumption does not reach completion, the net result of these processes is a pronounced ^{15}N -enrichment in the residual NO_3^- pool (Voss et al., 2001) similar to that of the experimentally determined for heterotrophic denitrification ($\epsilon_{\text{NO}_3-\text{N}_2} \sim +15$ to $+30\text{‰}$) (Granger et al., 2008; Mariotti et al., 1981). When NO_3^- consumption proceeds to completion, as in anoxic porewaters, hardly any isotope effect is expressed (Lehmann et al., 2007). Finally, within stable redox transition zones, nitrate (from above) and ammonium (from below) are also near-quantitatively converted into N_2 or N_2O resulting in a discrete zone of ^{15}N -enrichment in nitrate and ammonium ($\epsilon_{\text{NO}_3-\text{N}_2} \sim +10\text{‰}$; $\epsilon_{\text{NH}_4-\text{N}_2} \sim +10\text{‰}$) tied to decreasing concentrations (Fuchsman et al., 2008; Thunell et al., 2004; Wenk et al., 2014).

The ocean's redox structure thus tightly controls the nitrogen cycle through the complex network of metabolic pathways by which nitrogen flows between reservoirs. The local redox conditions determine the dissolved inorganic nitrogen speciation, nitrate being stable in oxic waters and ammonium in anoxic waters. The nature (i.e. OMZs versus redox transition zones), location (shallow versus deep), and spatial extent of dysoxic zones determine the

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