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Molybdenum drawdown during Cretaceous Oceanic Anoxic Event 2

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ABSTRACT

During the Cretaceous greenhouse, episodes of widespread ocean deoxygenation were associated with globally occurring events of black shale deposition. Possibly the most pronounced of these oceanic anoxic events (OAE's) was the Cenomanian–Turonian OAE2 (∼94 Ma). However, although certain redox sensitive trace metals tend to be preferentially sequestered in sediments deposited under anoxic conditions, with Mo drawdown being specifically prone to euxinic settings, these elements are generally somewhat depleted in sediments deposited during OAE2. To understand the driving factors responsible for this depleted trace metal drawdown, we have studied a low latitude section from the proto-North Atlantic Ocean (Tarfaya S57), where existing biomarker and iron–sulphur data point to a dominantly euxinic water column, with periodic transitions to ferruginous (Fe-rich) water column conditions. We utilise a variety of redox proxies (Fe-speciation, redox sensitive trace metals and Mo isotopes), which, in combination, allows us to evaluate the detailed nature of ocean redox conditions and hence controls on trace metal drawdown. The results suggest that seawater δ^{98}Mo values may have ranged between ∼0.6 and 1.1‰ during OAE2, likely connected to changes in the local Mo reservoir as a consequence of low and probably heterogeneous concentrations of Mo in the ocean. The very low Mo/TOC ratios at Tarfaya and elsewhere in the proto-North Atlantic may support a model in which deep-water circulation was partially restricted within and between the North Atlantic and other ocean basins. We propose that the combination of a low and possibly heterogeneous δ^{98}Mo of seawater Mo, together with low Mo/TOC ratios, points to a large decrease in the global oceanic Mo reservoir during OAE2, reflecting a major global scale increase in Mo drawdown under persistent euxinic conditions.

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1. Introduction

The Cretaceous was a time of high global temperatures as a consequence of elevated atmospheric CO_2 (Friedrich et al., 2012). A series of major black shale deposition events during intervals of ocean deoxygenation (termed oceanic anoxic events; OAEs) were associated with these extreme greenhouse conditions (Jenkyns, 2010). These fluctuations in Cretaceous seawater redox conditions coincided with massive perturbations to the carbon and nutrient cycles within and between the main global reservoirs (Friedrich et al., 2012). During the Cenomanian–Turonian (Bonarelli) event (OAE2) at ∼95 Ma, a worldwide positive carbon isotope excursion documents the globally enhanced burial of marine organic carbon under anoxic conditions (Jenkyns, 2010), likely driven by a global-scale increase in primary productivity (Schlanger and Jenkyns, 1976; Kuypers et al., 2002; Mort et al., 2007).

Anoxic conditions favoured the widespread deposition of organic-rich black shales during and intermittently prior to OAE2, in the proto-North Atlantic and western Tethys Ocean (Kuypers et al., 2004; Lüning et al., 2004; Kolonic et al., 2005; Hetzel et al., 2009) and in other regions (Lenniger et al., 2014). Several studies have argued that sluggish circulation within the proto-North Atlantic and restricted deep-water exchange with other ocean basins (Friedrich et al., 2012; Lüning et al., 2004) was responsible for black shale deposition (Monteiro et al., 2013; Wagner et al., 2013). However, other studies have postulated that sufficient ocean circulation existed to allow rigorous water exchange with the surrounding ocean basins (Jiménez Berrocoso et al., 2010).

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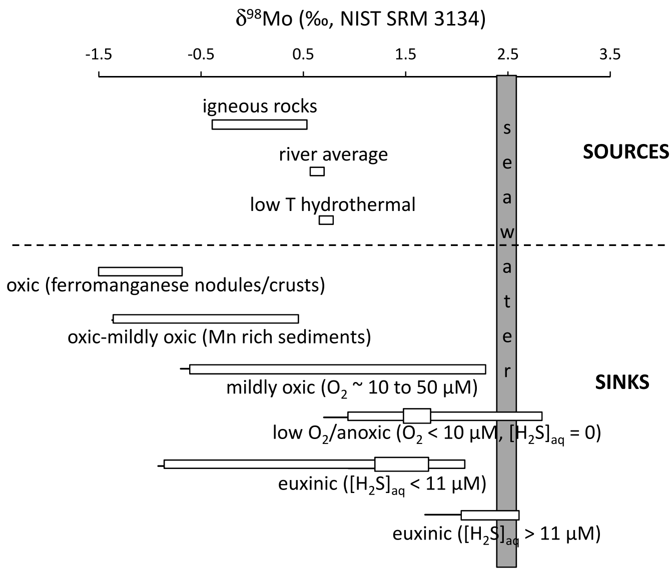

Fig. 1. The main oceanic Mo sources and sinks, and their isotopic compositions. Note that δ^{98}Mo has been renormalised to NIST SRM 3134, according to Goldberg et al. (2013). Narrow boxes represent the reported data range for each environment; wide boxes represent areas of data concentration. δ^{98}Mo values compiled from Vögelin et al. (2012 and references therein) and Goldberg et al. (2012, 2013, and references therein).

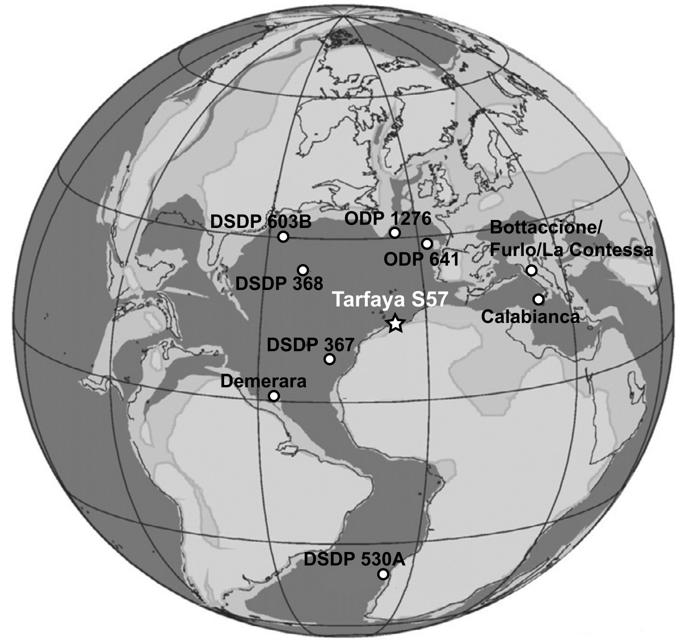

Fig. 2. Continental reconstruction for 94 Ma (modified from Forster et al., 2008), with the study site (Tarfaya S57) and other locations discussed in this study. Light grey areas represent land and middle grey areas represent flooded continental plates. Thin black lines outline present day coastlines.

Under anoxic conditions, many redox sensitive trace metals are preferentially deposited (Crusius et al., 1996; Algeo and Maynard, 2004), with Mo drawdown being specifically prone to euxinic (free H_2S) settings (Zheng et al., 2000). However, during the onset of OAE2 a severe depletion of sedimentary Mo, V, Zn, Cr, Ni and U (relative to total organic carbon; TOC) has been reported for the North Atlantic (Kolonic et al., 2005; Hetzel et al., 2009). This observation has been explained by a reduction in the marine trace element inventory due to the enhanced drawdown of redox sensitive elements through expansion of euxinic waters (Hetzel et al., 2009). However, low Mo/TOC ratios in euxinic sediments have also been suggested to be typical of silled marine basins, where resupply of Mo via deep water renewal was limited, resulting in lower oceanic Mo concentrations and hence less uptake of Mo per unit of organic matter compared to fully connected ocean basins (Algeo and Lyons, 2006; McArthur et al., 2008). This raises the question as to whether drawdown of Mo during OAE2 occurred on a local, regional or global scale, with consequences for identifying the global extent and severity of ocean anoxia during a period of massive, global carbon–climate perturbation.

Mo isotopes have emerged as a powerful tool for evaluating the global extent of ocean euxinia and the size of the oceanic Mo repository (Pearce et al., 2008; Dickson et al., 2012; Kendall et al., 2011; Westermann et al., 2014). The relative extent of oxic and anoxic sinks defines the amount and isotopic composition of Mo remaining in seawater (see supplementary information). The ability of Mo isotopes to capture the seawater signal is, however, limited to highly sulfidic environments, whereas under both oxic conditions and anoxic conditions where H_2S is absent or concentrations are low, a wide range of Mo isotope signatures may be encountered (Fig. 1).

This study focuses on a low latitude W-African shelf setting of the proto-North Atlantic Ocean (Tarfaya Basin). We present high resolution (∼0.3 ka scale) Mo isotope data that cover the initial onset and first part of the maximum perturbation of OAE2, as defined by the positive carbon isotope excursion (Kolonic et al., 2005; Tsikos et al., 2004). To determine the fidelity of sedimentary Mo isotopes to reflect seawater values, we critically appraise the Mo isotope composition of marine sediments with new and existing data that document local redox conditions (Fe-speciation, organic carbon contents, biomarkers and trace elements). Indeed, while the sediments were predominantly deposited under sulfidic conditions (Kolonic et al., 2005; Mort et al., 2008), regular cyclic (orbital timescale) transitions to anoxic, Fe-rich (ferruginous) water column conditions were also a prominent feature (Poulton et al., 2015). By combining hydrographic and geochemical aspects, our Mo isotope data provide a new view on euxinic water mass expansion and extensive Mo drawdown during OAE2.

1.1. The Late-Cretaceous Tarfaya shelf basin

The Tarfaya Basin is located on the northwestern margin of the Sahara Craton. During the Late-Cretaceous, the basin was situated at a latitude of ∼15°N in a marine shelf setting open to the proto-North Atlantic Ocean (Fig. 2) (Lüning et al., 2004; Kolonic et al., 2005; Wagner et al., 2013; Mort et al., 2008; Kuhnt et al., 2009). The N-African palaeo-coast is proposed to have been in the centre of an upwelling zone that covered large parts of the subtropical African margin and was responsible for providing nutrient-rich water and fostering the establishment of anoxia/euxinia (Lüning et al., 2004; Kolonic et al., 2005; Wagner et al., 2013; Mort et al., 2008).

Core samples were obtained from drill site S57 of the Tarfaya Basin, covering the core interval between 50.04 to 54.35 m. Palaeowater depths at site S57 have been estimated as ∼250 to 300 m (Kolonic et al., 2005). The section consists of pelagic organic-rich carbonates and marls, with 2 to 20 wt.% TOC and very little terrigenous input (Kolonic et al., 2005). Turbidites are absent from the studied part of the section. The last occurrence of *Rotalipora cushmani* (∼93.9 Ma) was found at ∼51.2 m (Tsikos et al., 2004). The studied section covers the upper δ^{13}C phase A to lower phase B with a positive δ^{13}C excursion from −27 to −23‰ (Fig. 3) (Kolonic et al., 2005). Spectral analyses, based largely on cyclicity in organic matter content, point to a dominance of obliquity forcing with a

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