

# Sulfur cycle imbalance and environmental change during the Ediacaran Period

## *Desequilibrio del ciclo del azufre y cambio ambiental durante el Período Ediacárico*

G.A. Shields<sup>1</sup>, B.J.W. Mills<sup>2</sup>

<sup>1</sup>Department of Earth Sciences, University College London, Gower Place, London, WC1E 6BT, UK. Email: [g.shields@ucl.ac.uk](mailto:g.shields@ucl.ac.uk); ORCID ID: <https://orcid.org/0000-0002-7828-3966>

<sup>2</sup>School of Earth and Environment, University of Leeds, Leeds, LS2 9JT, UK. ORCID ID: <https://orcid.org/0000-0002-9141-0931>

### ABSTRACT

A different approach is proposed here to solve the problem of negative  $\delta^{13}\text{C}$  excursions during the Ediacaran, by viewing them in terms of a linked carbon-sulfur-oxygen system, whereby changes in oxidant dynamics caused an excess of organic carbon oxidation over burial, resulting in a smaller DOM reservoir. The amount of oxidant required to achieve a deep negative carbon isotope excursion through net organic carbon oxidation may reasonably result from basin-scale evaporite dissolution.

**Keywords:** Carbon isotopes; Sulfur isotopes; Biogeochemical balance; Ediacaran.

### RESUMEN

Se propone aquí un enfoque diferente para resolver el problema de las excursiones quimioestratigráficas negativas durante el Ediacárico, considerándolas en términos de un sistema vinculado de carbono-sulfuro-oxígeno, en el que los cambios en la dinámica de los oxidantes causarían un exceso de oxidación de carbono orgánico sobre el enterramiento, lo que resultaría en un depósito menor de DOM. La cantidad de oxidante requerida para lograr una excursión isotópica de carbono negativa a través de la oxidación de carbono orgánico neto puede resultar razonablemente de la disolución de evaporitas a escala de cuenca.

**Palabras clave:** Isótopos de carbono; Isótopos de azufre; Balance biogeoquímico; Ediacárico.

### Introduction

The Ediacaran Period (ca. 635-540 Ma) marks a turning point in Earth history when groups of morphologically complex multicellular eukaryotes, including algae and animals, attained ecological

dominance, irrevocably changing Earth System dynamics (Lenton *et al.*, 2014). These biological radiations took place after two prolonged episodes of global glaciation during the Cryogenian Period (ca. 715-660 and ca. 650-635 Ma), and were accompanied by further short-lived (ca. 580 Ma

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and ca.550 Ma) regional ice ages, interspersed with warmer intervals (Condon *et al.*, 2005; Pu *et al.*, 2016; Le Heron *et al.*, 2019). The world's oceans also became episodically more oxygenated during the Ediacaran with the extent of oxygenated seafloor reaching near-modern levels at times by the early Cambrian (Chen *et al.*, 2015). Both climate and oxygenation are regulated by Earth's long-term carbon cycle, and so perhaps unsurprisingly this interval is characterised by extreme carbon isotope instability (Kaufman *et al.*, 1997). Since its discovery over 30 years ago (Burns & Matter, 1993; Calver, 2000), the uniquely high amplitude of the Shuram  $\delta^{13}\text{C}$  anomaly has led some to question conventional interpretations of carbonate carbon isotopes (Melezhik *et al.*, 2005; Schrag *et al.*, 2011). However, its pivotal position beneath fossil evidence for the first biomineralising, bioturbating and bilaterian animals encourages us to venture a more satisfying, parsimonious explanation for the Shuram anomaly, one that ties together carbon cycle perturbations with life's evolution.

Conventional carbon isotope mass balance is based on the principle that the isotopic composition of carbon input via outgassing and weathering, and that of sedimentary carbon outputs are equal on time scales of  $>10^5$  years (Garrels & Lerman, 1984). This  $\delta^{13}\text{C}$  value is considered to be unchanging at -5‰: the average composition of crustal carbon. Because organic matter is depleted in  $^{13}\text{C}$ , and carbonate rocks precipitate in isotopic equilibrium with ambient dissolved inorganic carbon (DIC), the mean  $\delta^{13}\text{C}$  value of carbonate rocks and fossils can be used to determine the proportion that sedimentary organic matter makes up of the total sedimentary carbon sink. This proportion is generally referred to as  $f_{\text{org}}$ , which has varied over Earth history between 0.1 and 0.3 (Krissansen-Totton *et al.*, 2015). Using a conventional mass balance approach, extreme negative anomalies can only be explained by negative rates of organic burial. This is particularly true of the late Ediacaran Shuram excursion when  $\delta^{13}\text{C}$  remained below -8‰ for at least  $\sim 10$  Myr (Condon *et al.*, 2005; Gong *et al.*, 2017). In order to address this quandary, it was proposed that the pool of dissolved organic matter (DOM) in the Proterozoic ocean was much larger than today, and that negative excursions represent non-steady-state remineralisation of that pool (Rothman *et al.*, 2003). However, later numerical

treatments of this model pointed out that the Earth system cannot remain out of oxygen (and carbon) balance for such a long period of time (Bristow & Kennedy, 2008). In other words, there is insufficient oxidant even in the modern atmosphere and oceans to remineralize enough organic matter to drive a -8‰  $\delta^{13}\text{C}$  excursion for several million years. As a result, many authors have interpreted extreme negative anomalies as diagenetic artefacts, biased sampling of authigenic cements or as purely regional phenomena (Bristow & Kennedy, 2008; Derry, 2010; Grotzinger & Fike, 2011; Schrag *et al.*, 2013; Li *et al.*, 2017; Shi *et al.*, 2018). However, such arguments appeal to an inexplicable sampling bias, whereby globally correlative isotopic signatures are presumed to be unrepresentative of the global carbonate sink.

Here we take a different approach to the problem of negative  $\delta^{13}\text{C}$  excursions by viewing them in terms of a linked carbon-sulfur-oxygen system, whereby changes in oxidant dynamics caused an excess of organic carbon oxidation over burial, resulting in a smaller DOM reservoir. For steady state to be maintained throughout a negative  $\delta^{13}\text{C}$  excursion, shrinkage of the DOM pool would need to match surplus oxidant production for the duration of the anomaly. If we consider plausible  $\delta^{13}\text{C}$  values of -10‰ and -35‰ for deposited carbonates and kerogen (globally averaged carbon sinks), respectively, and -30‰ and -5‰ for the DOM reservoir and crustal carbon (globally averaged carbon sources), respectively (Lu *et al.*, 2013; Krissansen-Totton *et al.*, 2015; Lee *et al.*, 2015), then organic carbon oxidation would need to increase over the background rate by approximately a factor of three. In other words, and following conventional C-isotope mass balance [ $\delta^{13}\text{C}_{\text{carb}} = (\delta^{13}\text{C}_{\text{carb-org}}) \cdot f_{\text{org}} + \delta^{13}\text{C}_{\text{input}}$  at steady state], when  $f_{\text{org}} = 0.1$ ,  $\delta^{13}\text{C}_{\text{input}} = -12.5$ ‰ and  $f_{\text{DOM}} = 0.3$ , whereby  $f_{\text{DOM}}$  is the proportion that marine organic matter oxidation contributes to the overall carbon flux. The oxidant imbalance ( $f_{\text{DOM}} = 0.3$ ) would have needed three times as much oxygen as could have been supplied by organic burial ( $f_{\text{org}} = 0.1$ ) alone, and so requires a contribution from other sources, most likely pyrite burial.

Although bacterial sulfate reduction coupled with pyrite burial releases on a mole-for-mole basis almost twice as much oxygen as organic burial, it is generally assumed that the oxygen released by

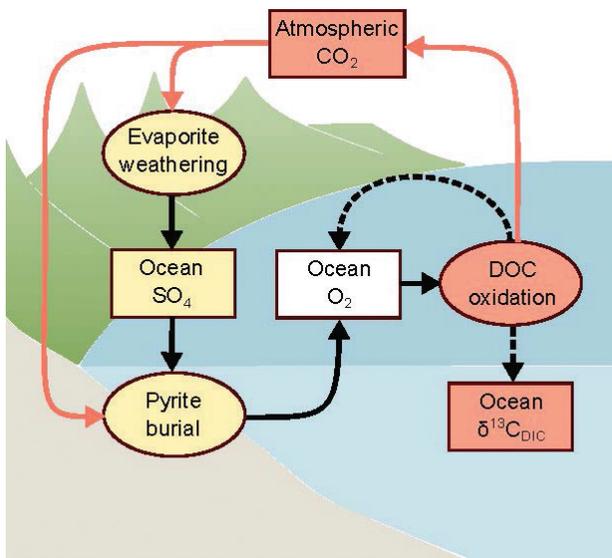


Figure 1.—Feedback diagram illustrating the effects of evaporite weathering on ocean oxygenation and  $\delta^{13}\text{C}$ . Boxes show quantities, ovals show processes, whilst yellow indicates sulfur cycle and red indicates carbon cycle. An evaporite dissolution event results in a large flux of sulfate to the ocean, permitting high rates of pyrite burial, which increases atmospheric and ocean  $\text{O}_2$ . As the deep ocean becomes oxygenated, depletion of the dissolved organic carbon (DOC) reservoir represents a negative feedback on ocean oxygenation and drives ocean  $\delta^{13}\text{C}$  to negative values. Red arrows show potential for positive feedback: DOC oxidation increases atmospheric  $\text{CO}_2$ , leading to higher surface temperature and greater rates of precipitation and runoff, which fuels further evaporite dissolution and nutrient delivery. Solid arrows show positive effects and dashed arrows show negative effects.

pyrite burial is approximately matched by the oxygen consumed during pyrite weathering. However, most riverine sulfate derives from the weathering of evaporites (Burke *et al.*, 2018), rates of which due to the sporadic and regional nature of evaporite deposits, will vary considerably over time (Wortmann & Paytan, 2012). During parts of the Proterozoic, when oceans were both iron-rich and anoxic, and so prone to sulfate reducing conditions (euxinia) at productive margins, one might realistically suppose that the amount of oxidising power transferred from rock sulfate to the surface environment would also have varied considerably, particularly during times when no basin-scale evaporite deposits were forming. We refer to this as ‘sulfur cycle imbalance’, meaning that the oxidant required to oxidise pyrite S was not balanced by the oxidant released by pyrite S burial and/or that the  $\text{SO}_4$  flux to the ocean from evaporite

weathering was not balanced by evaporite deposition (on timescales over which the long-term carbon and oxygen cycles must be in balance).

By writing a simple steady state mass-balance for the surface carbon cycle we can map the broad relationship between the evaporite sulfate input rate, net DOM oxidation and  $\delta^{13}\text{C}$  composition of the ocean-atmosphere system. These calculations show that an evaporite weathering flux of around  $1.1 \times 10^3 \text{ mol S yr}^{-1}$  (around 10 times the modern flux, as proposed for the dissolution event during the early Cenozoic could sustain a carbon isotope excursion of between -10‰ and -15‰, depending on the proportion of the riverine sulfate flux that is eventually buried as pyrite. Thus, the amount of oxidant required to achieve a deep negative carbon isotope excursion through net organic carbon oxidation may reasonably result from basin-scale evaporite dissolution.

### Sulfur cycle imbalance during the Ediacaran

The second half of the Tonian Period (ca. 830–770 Ma) witnessed the largest evaporite depositional event of pre-Ediacaran time during the break-up of the supercontinent Rodinia (Evans, 2006; Turner & Bekker, 2016; Schmid, 2017). Estimates for the current preserved volume of Tonian evaporites range from 375,000  $\text{km}^3$  to 912,400  $\text{km}^3$ , which places Tonian deposits among the largest basin-scale evaporite deposits ever (Evans, 2006; Prince *et al.*, 2019). They include: the Centralian Superbasin, Australia (683,000  $\text{km}^3$ ); Amundsen and Mackenzie-Ogilvie basins, Canada (193,000  $\text{km}^2$ ); Adelaide fold belt, South Australia (100,000  $\text{km}^2$ ); Duruchaus basin, Namibia (30,000  $\text{km}^2$ ); and the Central African copperbelt (50,000  $\text{km}^2$ ). Despite diverse geodynamic settings, these Tonian evaporite basins underwent inversion during middle to late Ediacaran time, mostly after ca. 570 Ma, and broadly coincident with the ‘Pan-African orogeny’ or ‘Transgondwanan supermountain’ events (Campbell & Squire, 2010). Specifically, Centralian evaporites were exhumed by deformation associated with the Kuunga Orogen; Amundsen-Mackenzie-Ogilvie evaporites were exhumed on a passive margin; South Australian evaporites were exhumed during development of a raised volcanic rift margin; Duruchaus was exhumed

by early Damaride accretion of terranes onto an active margin; and the Copperbelt was exhumed by intrusion of an anorogenic large igneous province. Subsequent erosion means that initial volumes must have been substantially higher. The high sulfate mineral content of these evaporite deposits (Fakhraee *et al.*, 2019) and relatively unchanging seawater  $\delta^{34}\text{S}$  values (Strauss, 1993) are consistent with the build-up of a large marine sulfate reservoir during the early Neoproterozoic. Sulfur cycle imbalance (excess sulfate weathering over deposition, coupled with excess pyrite deposition over weathering) is likely to have continued until massive evaporite deposition began in the ‘Palaeotethyan’ realm after c.550 Ma (Schroder *et al.*, 2004), at which point the imbalance seems likely to have been reversed.

### Environmental effects of sulfur cycle imbalance

This evaporite dissolution / DOM oxidation scenario appears to be the most parsimonious solution to the Shuram C-isotope conundrum in that it predicts the extent of oxic seafloor to increase towards the end of the excursion (Chen *et al.*, 2015; Kendall *et al.*, 2015), while maintaining high sulfate concentrations, which is in line with geochemical studies (Shi *et al.*, 2018). Net oxidation of organic carbon (via pyrite burial) should cause a substantial rise in atmospheric  $\text{CO}_2$  concentration, leading to global warming and enhanced continental weathering, with the potential to drive further evaporite dissolution and therefore sustain oxidant delivery.

Although our model fits best the late Ediacaran Shuram anomaly, sulfur cycle imbalance may have also played a role in other carbon isotope excursions. The interval of major carbon cycle disruption from ca.770 Ma until ca.550 Ma was a time of little or no basin-scale evaporite deposition (Guilbaud *et al.*, 2015), suggesting that, as in the Cenozoic, the sulfate weathering-deposition cycle was not in steady-state. However, unlike the Cenozoic, the low atmospheric oxygen and anoxic deep ocean of the Neoproterozoic allowed evaporite-derived oxidizing power to be effectively transmitted into a negative  $\delta^{13}\text{C}$  signal. Such imbalance, potentially driven by weathering events and related positive feedbacks, may be particularly relevant to the ending

and immediate aftermath of ‘Snowball Earth’ glaciations, which are marked by euxinia and anomalously high levels of pyrite deposition (Sahoo *et al.*, 2012; Lang *et al.*, 2018).

The existence of a series of negative carbon isotope excursions during much of the Neoproterozoic Era indicates that the DOM pool underwent dynamic size changes throughout this time and served as a buffer against oxygenation and climate change, but only when the pool was sufficiently large. The absence of a large DOM buffer in the ocean, climate and oxygen levels would have been rendered more vulnerable to change. There are tantalising hints that this may have been the case after c.550 Ma (Tostevin *et al.*, 2019), and that the ocean redox balance remained sensitively balanced throughout the Ediacaran-Cambrian transition interval (He *et al.*, 2019). If exhaustion of the DOM pool occurred during the Shuram anomaly, it is plausible to suppose that the subsequent expansion of aerobic Ediacaran fauna was an opportunistic radiation in response to a transient oxidant surplus.

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