

Persistent dysoxia in very shallow seas across the late Cambrian SPICE event, Durness Group, UK

Ke Feng¹, Fred Bowyer^{1,2}, Andrew Curtis¹, Simon W. Poulton², Laetitia Pichevin¹, and Rachel Wood^{1,*}

¹School of GeoSciences, University of Edinburgh, Edinburgh EH9 3FE, UK

²School of Earth and Environment, University of Leeds, Leeds LS2 9JT, UK

ABSTRACT

The global late Cambrian Steptoean Positive Isotopic Carbon Excursion (SPICE; ca. 495–492 Ma) has been linked to oceanic anoxia followed by a pulse of atmospheric oxygenation that in turn may have facilitated the Ordovician Radiation. To provide new insight into the evolution of very-shallow-water redox conditions across the SPICE, we present an integrated multiproxy study of carbonate rocks from the Durness Group, Scotland, UK, combining Fe speciation, redox-sensitive trace element systematics, and I/(Ca + Mg) ratios. We interpret the Durness SPICE peak to have occurred during an early highstand interval, where shallow waters were impacted by either regional mobilization of Fe²⁺ under low-oxygen conditions or episodic upwelling of deep, ferruginous, anoxic waters that followed high-frequency cycles. Unlike other records, our data show that very low marine oxygen concentrations (dysoxia) persisted before, during, and after the SPICE, confirming that a very shallow redoxcline was maintained in this region, with no evidence for any sustained increase in oxygenation. We conclude that under the prevailing low-atmospheric-oxygen conditions of the Cambrian, dysoxia was prevalent even in very shallow waters.

INTRODUCTION

The Steptoean Positive Isotopic Carbon Excursion (SPICE) was a notable global event marked by a >10‰ positive carbon isotope ($\delta^{13}\text{C}$) excursion (CIE) during the late Cambrian (ca. 495–492 Ma; Saltzman et al., 2000; Cothren et al., 2022). The onset of the SPICE broadly coincides with a major biotic turnover of trilobites (e.g., Yang et al., 2024). The rising limb of the SPICE has been linked to a global deoxygenation event characterized by an expansion of low-oxygen oceanic conditions and increased organic carbon and pyrite burial (Saltzman et al., 2000), which occurred against a backdrop of generally low sulfate concentrations (Gill et al., 2011).

The SPICE onset is characterized by the replacement of shallow-water taxa by those adapted to cold and deep-water environments and is accompanied by oxygen isotope evidence that suggests upwelling of colder deep waters onto shallow shelves (e.g., Elrick et al., 2011).

The same oxygen isotope data set also indicates a gradual increase in seawater temperature through the rising limb of the SPICE, potentially linked to decreasing thermohaline circulation and a temperature-related decrease in seawater oxygen concentrations (Elrick et al., 2011). The long-term burial of organic carbon and pyrite associated with the rising limb of the SPICE, in turn, is thought to have led to an increase in atmospheric $p\text{O}_2$ (Saltzman et al., 2011), so promoting an increase in phytoplankton diversity and, ultimately, potentially facilitating the Ordovician Radiation (e.g., Saltzman et al., 2011).

In Laurentian sections, the SPICE peak occurs approximately coincident with maximum regression near the Sauk II–Sauk III supersequence boundary (e.g., Saltzman et al., 2004; Cothren et al., 2022), suggesting a link to tectonic changes and thus continental weathering (Pulsipher et al., 2021), but there are different interpretations of sea-level change coincident with the SPICE (e.g., Schiffbauer et al., 2017; Yang et al., 2024). The trigger for the SPICE, however, remains elusive, given that mercury and osmium concentrations do not support


enhanced volcanism or weathering controls but instead indicate weak carbon-cycle feedbacks associated with inefficient silicate weathering (Frieling et al., 2024).

The shallow-marine Durness Group, NW Scotland, UK, exposes extensive carbonates that formed on a low-angle ramp as part of the vast tropical-subtropical carbonate bank of SE Laurentia (Fig. 1). Strata of the Eilean Dubh Formation archive the SPICE and are composed exclusively of very shallow limestone lithologies with evaporite pseudomorphs, with abundant evidence of frequent exposure within a sabkha, arid tidal-flat setting (see the Supplemental Material¹). Aeolian silts (the only source of clastics) are dispersed throughout and in a few places form thin beds capping multiple high-frequency cycles (described in Raine, 2010; Raine and Smith, 2012). There is no evidence for bioturbation and only a very limited skeletal fossil record. Glauconite and Hg enrichments have been interpreted to track low oxygen levels coincident with the SPICE, followed by a transient, local oxygenation event (Pruss et al., 2019).

Here, we combine major element concentration data with multiple paleoredox proxy data, including Fe speciation, redox-sensitive trace element (RSTE) systematics, and I/(Ca + Mg) ratios, to further constrain the dynamics of the SPICE event and its aftermath. Specifically, our multiproxy approach allows a detailed assessment of the redox evolution of a very shallow depositional setting, as documented by the Durness Group, providing new insight into global-scale variability in shallow-water redox conditions under the low-atmospheric-oxygen conditions that characterized the late Cambrian.

METHODS

Carbonate powders from micritic carbonates and grainstones were microdrilled and analyzed

Rachel Wood  <https://orcid.org/0000-0002-0165-5987>
*Rachel.Wood@ed.ac.uk

¹Supplemental Material. Sedimentology and stratigraphy, methods, and data tables. Please visit <https://doi.org/10.1130/GEOL.S.28851395> to access the supplemental material; contact editing@geosociety.org with any questions.

CITATION: Feng, K., et al., 2025, Persistent dysoxia in very shallow seas across the late Cambrian SPICE event, Durness Group, UK: *Geology*, v. 53, p. 642–646, <https://doi.org/10.1130/G52950.1>

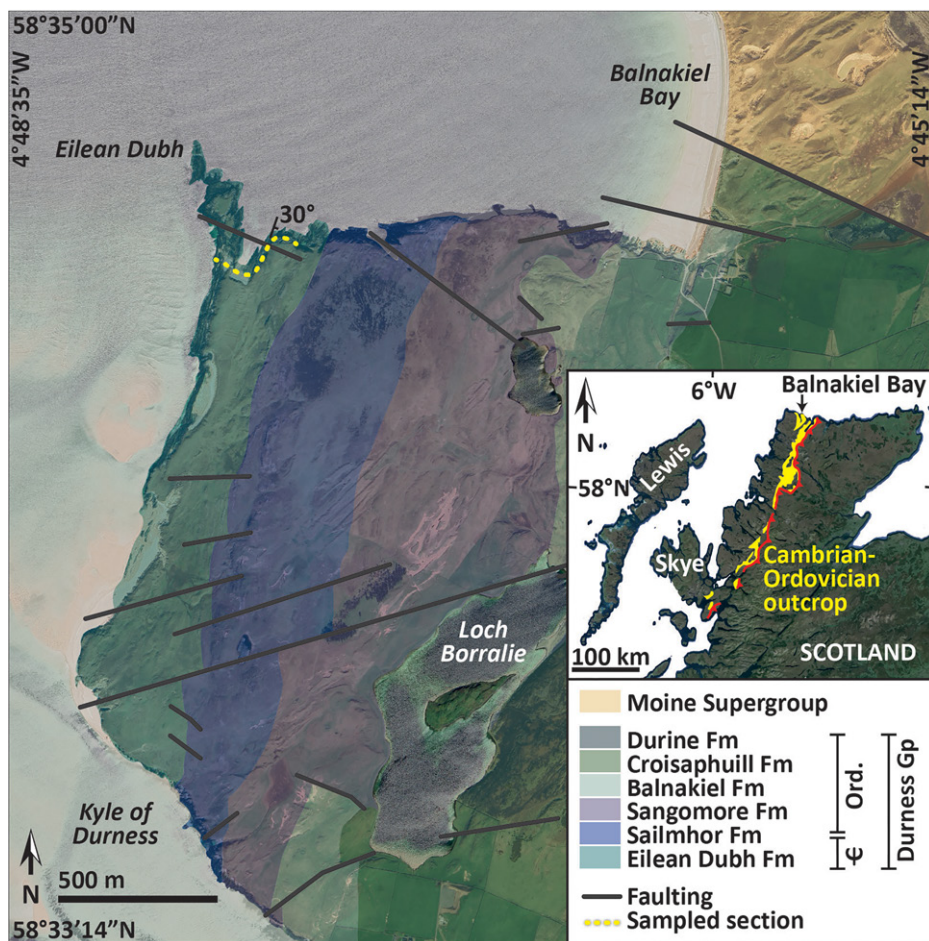


Figure 1. Geological map of Cambrian–Ordovician Durness Group, NW Scotland, UK, showing study area southwest of Balnakeil Bay, Durness (modified after Raine, 2010).

for $\delta^{13}\text{C}_{\text{carb}}$ and $\delta^{18}\text{O}_{\text{carb}}$ (carbonate) using continuous flow isotope ratio mass spectrometry at Isoanalytical Laboratories (Crew, UK). Reproducibility of standards was 0.05‰ for $\delta^{13}\text{C}$ and 0.1‰ for $\delta^{18}\text{O}$. Iron speciation analyses (Poulton and Canfield, 2005), major element concentrations using inductively coupled plasma–optical emission spectroscopy (ICP-OES), and trace element concentrations using inductively coupled plasma–mass spectrometry (ICP-MS) were all undertaken at the Cohen Laboratories, University of Leeds. Measurements of $\text{I}/(\text{Ca} + \text{Mg})$ ratios (Lu et al., 2018) were undertaken using ICP-OES and ICP-MS at the University of Edinburgh, using a tellurium internal standard (see the Supplemental Material for further details of methods and redox proxy systematics and Tables S1 and S2 for all data).

RESULTS AND DISCUSSION

We interpret the study interval to represent the end of the Sauk IIA to the beginning of the Sauk IIIA supersequences (Fig. 2A). Our $\delta^{13}\text{C}_{\text{carb}}$ data are consistent with previous records (Pruss et al., 2019), whereby the SPICE onset and rising limb occur within the transgression to highstand

of Sauk IIB, reaching a maximum $\delta^{13}\text{C}$ value of +2.78‰ (Fig. 2B). Maximum regression across Sauk II–Sauk III is recorded near the SPICE peak (but see the Supplemental Material).

Samples through the entire interval prior to the SPICE have iron concentrations <0.32 wt% (Fig. S1 in the Supplemental Material), combined with very low total organic carbon (TOC) contents (<0.03 wt%; Fig. 2C). Iron concentrations then rise to 0.42 wt% just prior to the onset of the SPICE and become highly variable, from 0.15 wt% to 1.26 wt%, following meter-scale sedimentary cycles (Fig. 2A). TOC concentrations also follow meter-scale sedimentary cycles and reach a maximum (but still <0.1 wt%) at the maximum relative water depth (the maximum flooding surface) of Sauk IIB.

Samples with iron concentrations >0.5 wt% (Clarkson et al., 2014) were analyzed for total iron/aluminum ($\text{Fe}_\text{T}/\text{Al}$), highly reactive/total iron ($\text{Fe}_{\text{HR}}/\text{Fe}_\text{T}$), and pyrite-bound iron/highly reactive iron ($\text{Fe}_{\text{py}}/\text{Fe}_{\text{HR}}$) ratios (Figs. 2D–2F; Table S2). Samples with $\text{Fe}_\text{T} > 0.5$ wt% and $\text{Al} > 0.5$ wt% have $\text{Fe}_\text{T}/\text{Al}$ ratios ranging from 0.33 to 2.44, with seven samples yielding $\text{Fe}_\text{T}/\text{Al} > 0.66$, indicating Fe enrichment (Fig. 2D; Clarkson et al., 2014).

Samples are also characterized by $\text{Fe}_{\text{HR}}/\text{Fe}_\text{T}$ ratios that are persistently elevated above the anoxic threshold ($\text{Fe}_{\text{HR}}/\text{Fe}_\text{T} > 0.38$; Fig. 2E; Poulton and Canfield, 2011), with $\text{Fe}_{\text{py}}/\text{Fe}_{\text{HR}}$ ratios that are depleted relative to the lower limit for possible euxinia ($\text{Fe}_{\text{py}}/\text{Fe}_{\text{HR}} < 0.60$; Fig. 2F; Poulton, 2021). The common occurrence of elevated $\text{Fe}_{\text{HR}}/\text{Fe}_\text{T}$ and $\text{Fe}_\text{T}/\text{Al}$ ratios across the SPICE indicates at least periodic anoxic mobilization of Fe^{2+} and subsequent precipitation, while low $\text{Fe}_{\text{py}}/\text{Fe}_{\text{HR}}$ ratios suggest limited production of sulfide, even during diagenesis.

To provide further insight into redox conditions at the site of deposition, we use a refined approach for calculating enrichment factors (termed EF^*) for redox-sensitive trace metals (e.g., U, Mo, Re) in carbonate-rich lithologies (Krewer et al., 2024; Li et al., 2025; see the Supplemental Material). The resultant EF^* values for U and Mo are generally low (Figs. 2G–2H; Table S2), indicating limited enrichment under anoxic water-column conditions (Tribouillard et al., 2012; Li et al., 2025). However, a cross-plot of Mo_{EF^*} versus U_{EF^*} (Fig. 3) reveals two pathways for the samples displaying RSTE enrichment, with the majority of elevated Mo_{EF^*} values resulting from delivery to sediments via a particulate shuttle mechanism (Tribouillard et al., 2012).

We further measure $\text{I}/(\text{Ca} + \text{Mg})$ ratios to potentially reveal more subtle variations in oxygen availability through time (Fig. 2J; Table S2). $\text{I}/(\text{Ca} + \text{Mg})$ ratios are very low throughout the section (0.17–0.4 $\mu\text{mol}/\text{mol}$, mean = 0.24 $\mu\text{mol}/\text{mol}$) relative to the typical “oxic” threshold of $\sim 2.6 \mu\text{mol}/\text{mol}$ (Glock et al., 2014; Lu et al., 2016), including prior to, during, and after the SPICE (Fig. 2J). These low values suggest limited iodide oxidation within the water column at the depth of carbonate formation (Lu et al., 2016). Calcite- and dolomite-bound iodine can be susceptible to diagenetic alteration (Hardisty et al., 2017), but the absence of any correlation between the $\text{I}/(\text{Ca} + \text{Mg})$ ratios and TOC, $\delta^{18}\text{O}$, and Mg/Ca (Fig. S2) argues against any significant influence from diagenetic alteration or organically bound iodine.

The iodine data suggest low but nonzero oxygen concentrations, conducive to iodate reduction (i.e., the water column was likely predominantly manganous to nitrogenous). Carbonate $\text{I}/(\text{Ca} + \text{Mg})$ ratios of <2.5 $\mu\text{mol}/\text{mol}$ (equivalent to $\text{IO}_3^- < 0.25 \mu\text{mol}/\text{L}$ in seawater) indicate seawater $[\text{O}_2]$ of <20–70 μM in modern and ancient oceans (Lu et al., 2016), suggesting that dysoxic conditions were prevalent at the site of carbonate formation throughout the Eilean Dubh Formation, consistent with the lack of large enrichments in U and Mo (Fig. 2), which would require water column anoxia. Significantly, there is no increase in $\text{I}/(\text{Ca} + \text{Mg})$ during or after the SPICE, but there is a broad transition from a lower interval (0–38.8 m, $n = 24$) where $\text{I}/(\text{Ca} + \text{Mg})$ values exhibit a higher mean (0.25

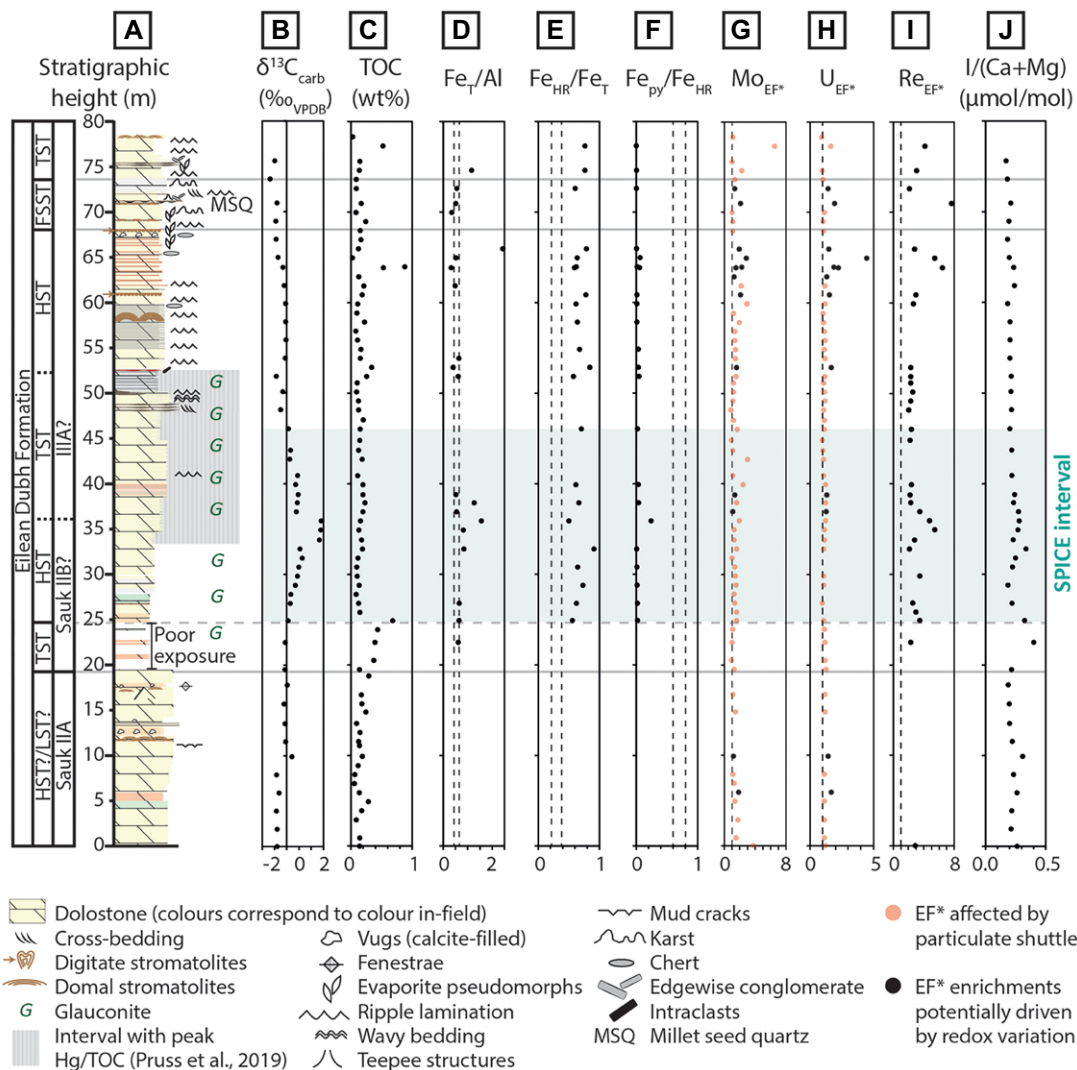


Figure 2. Redox dynamics through the late Cambrian Steptoean Positive Isotopic Carbon Excursion (SPICE) event (shaded in green), Eilean Dubh Formation. (A) Stratigraphy and correlation with Sauk supersequences (modified from Raine, 2010). Horizontal gray dashed line is maximum flooding surface. LST—lowstand systems tract; TST—transgressive systems tract; HST—highstand systems tract; FSST—falling stage systems tract. (B) $\delta^{13}\text{C}_{\text{carb}}$ record (this study). VPDB—Vienna Pee Dee belemnite. (C) Total organic carbon (TOC). (D) Fe_T/Al ratios (Fe_T —total iron) for samples with >0.5 wt% Fe, and >0.5 wt% Al. Vertical dashed lines bracket range of normal oxic values (0.44–0.66) (Clarkson et al., 2014). (E) $\text{Fe}_{\text{HR}}/\text{Fe}_T$ ratios (Fe_{HR} —highly reactive iron), where vertical dashed lines represent empirically derived “oxic” (<0.22) and “anoxic” (>0.38) threshold ratios, separated by an equivocal zone (Poulton and Canfield, 2011). (F) $\text{Fe}_{\text{py}}/\text{Fe}_{\text{HR}}$ ratios (Fe_{py} —pyrite-bound iron), where vertical dashed lines represent empirically derived ferruginous (<0.60) and euxinic (>0.80) thresholds, separated by an equivocal zone that may represent euxinia (Poulton, 2021). (G) Mo enrichment factors (Mo_{EF^*}). (H) U enrichment factors (U_{EF^*}). (I) Re enrichment factors (Re_{EF^*}). (J) $\text{I}/(\text{Ca} + \text{Mg})$.

$\mu\text{mol/mol}$) and higher standard deviation (0.05 $\mu\text{mol/mol}$) to an upper interval (40.9–75.6 m, $n = 18$) that exhibits a slightly lower mean (0.21 $\mu\text{mol/mol}$) and lower standard deviation (0.02 $\mu\text{mol/mol}$). Marginally elevated $\text{I}/(\text{Ca} + \text{Mg})$ values are largely restricted to two

horizons in the lower interval, prior to the SPICE (~20–25 m during the transgressive systems tract [TST] of Sauk IIB, and a short-lived peak represented by a single sample at 32.8 m, during the highstand systems tract [HST] of Sauk IIB; Fig. 2J).

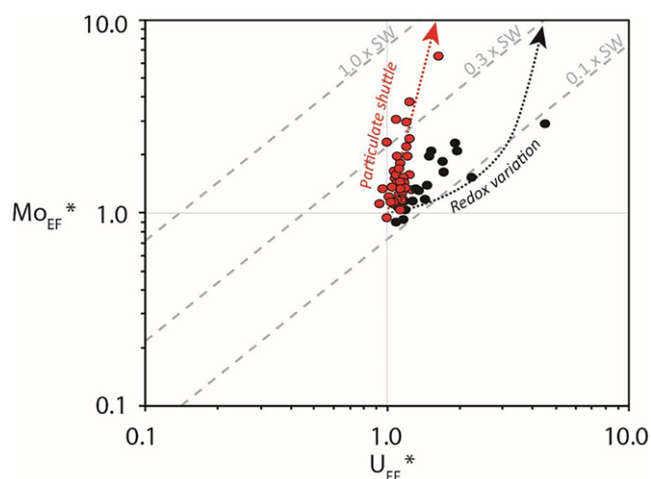


Figure 3. Cross-plot of revised Mo and U enrichment factors (EF^*) for carbonate samples of Eilean Dubh Formation. SW—sea water.

Intervals of elevated Re_{EF^*} (max = 7.57), in the general absence of co-enrichments in U (Fig. 2I), support the dysoxic bottom-water conditions indicated by the low but nonzero iodine data (Li et al., 2025). However, some samples higher in the section (~62–66 m) record appreciable U_{EF^*} values (Fig. 2H), likely corresponding to the short-lived development of anoxic conditions at the sediment–water interface, which is consistent with elevated TOC (Fig. 2C) and muted $\text{I}/(\text{Ca} + \text{Mg})$ ratios (Fig. 2J).

The strong evidence for dysoxic conditions throughout most of the section, along with evidence for the operation of a particulate shuttle that sequestered Mo in the sediment (Fig. 3), suggests that the observed enrichments in $\text{Fe}_{\text{HR}}/\text{Fe}_T$ reflect water-column mobilization of Fe^{2+} under regional anoxic conditions rather than an elevated source from intense continental weathering (see Wei et al., 2021). This is supported by the occurrence of elevated Fe_T/Al ratios in parts of the section (Fig. 2), given that both Fe_T and Al are generally con-

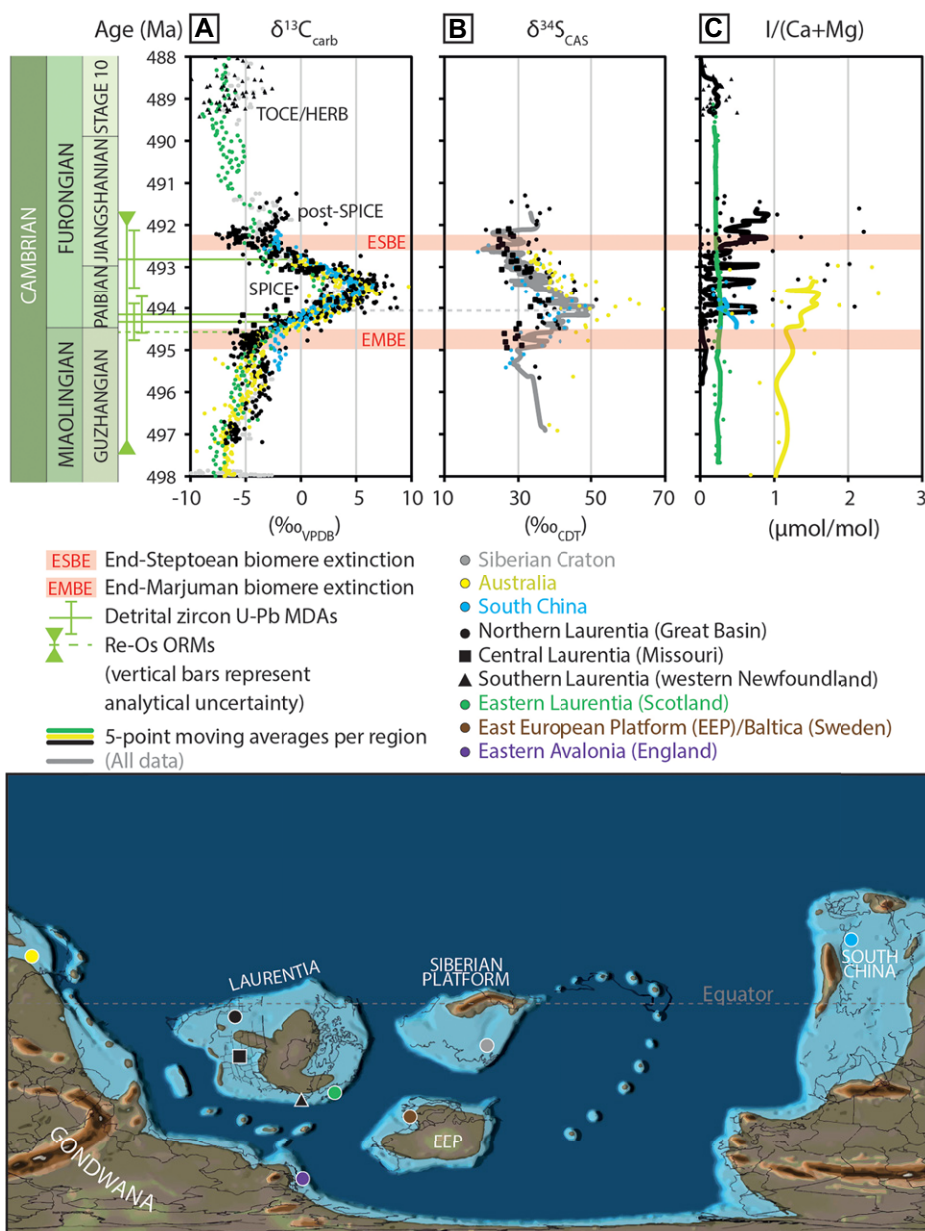


Figure 4. Global and regional geochemical expression of Steptoean Positive Isotopic Carbon Excursion (SPICE) event within a common age model and late Cambrian paleogeography for $\delta^{13}\text{C}_{\text{carb}}$ (A), $\delta^{34}\text{S}_{\text{CAS}}$ (carbonate associated silicate) (B), and $I/(\text{Ca} + \text{Mg})$ (C) (Gill et al., 2011; He et al., 2024; Pruss et al., 2019; Zhang et al., 2024). Biomere extinctions from Zhang et al. (2024); detrital zircon U-Pb data from Cothren et al. (2022); Re-Os age from Rooney et al. (2022); paleo-map output from Scotese (2021). VPDB—Vienna Pee Dee belemnite; V-CDT—Vienna Canyon Diablo Troilite; MDAs—maximum depositional ages; ORMs—organic-rich mudrocks; HERB—*Hellnmaria*–Red Tops Boundary; TOCE—top of Cambrian Excursion.

sidered immobile during chemical weathering (Canfield, 1997), and instead, elevated Fe_T/Al ratios reflect water-column drawdown of mobilized Fe^{2+} (Raiswell et al., 2018). Thus, taken together, our combined multiproxy paleoredox data indicate dominantly dysoxic conditions at the site of deposition, with persistent enrichments in Fe_{HR} being due to oxidation of Fe^{2+} sourced from either regional sediment-to-water mobilization under anoxic conditions or upwelling of ferruginous deeper waters.

COMPARISON WITH COEVAL RECORDS ACROSS THE SPICE

In the Durness Group, we interpret the onset of the SPICE rising limb to correspond with a TST and the peak of the SPICE with the HST, which is consistent with the overall pattern of sea-level change recorded in sections in the United States (Saltzman et al., 2004). This stacking pattern is shared by most sections that record the SPICE and is also noted for many other positive CIEs, including during the Ediacaran Shuram and the lower Cambrian (Bowyer et al., 2024).

Existing records show that widespread anoxia, or a shallow redoxcline, was pervasive across the continental shelves of western Laurentia during the pre- and early-SPICE interval (He et al., 2024). However, the low values of $I/(\text{Ca} + \text{Mg})$ recorded here, which remain within a narrow range during the whole interval studied, indicate relatively invariable low-oxygen conditions and notably a shallow marine redoxcline throughout the entire pre- to post-SPICE interval. By contrast, far higher mean $I/(\text{Ca} + \text{Mg})$ values have been recorded from other regions, including sections of Australia and the Great Basin of Laurentia (Fig. 4). These sites also record increasing $I/(\text{Ca} + \text{Mg})$ values either coincident with or after the SPICE, interpreted to reflect increasing local marine (and possibly atmospheric) oxygen concentrations driven by progressive organic carbon and pyrite burial (Fig. 4) (He et al., 2024). Such an increase in atmospheric oxygen concentrations would be expected to impact the shallow-marine redox state directly, and so it is noteworthy that the very shallow setting represented by the Eilean Dubh Formation does not record clear evidence for this increase.

Shelf and slope oceanic waters around the Iapetan and Laurentian continental margins during the latest Cambrian to Early Ordovician were generally poorly oxygenated, with shallow redoxclines or expanded oxygen minimum zones (OMZs) (e.g., Kozik et al., 2023). The ambient greenhouse climate would have favored the expansion of OMZs (e.g., Elrick et al., 2011), and ocean circulation may also have created heterogeneous distribution of nutrients and redox conditions (e.g., LeRoy et al., 2021).

Our data confirm that, unlike all other shallow marine records of SPICE events observed in other margins of Laurentia (UK, USA), South China, and Australia, the shallow waters of SE Laurentia did not experience any significant post-SPICE rise in oxygen (Fig. 4). It is possible that this could be due to seawater cooling, stimulating global ocean circulation (Zhang et al., 2024), accentuating continental margin upwelling (Stouffer et al., 2006), augmenting local productivity, and thereby expanding OMZs (Zhang et al., 2024). Such cooling is consistent with upwelling at the onset of the rising limb of the SPICE during the TST.

CONCLUSIONS

The Durness Group preserves a very-shallow-marine carbonate record from SE Laurentia across the late Cambrian SPICE and highlights the significant regional expression of this event, closely coupled to global sea-level change. Multiproxy paleoredox data show that the Durness SPICE interval was characterized by the introduction of ferruginous, anoxic waters into dysoxic shallow waters. Significantly, we show that very-low-marine-oxygen conditions per-

sisted before, during, and after the SPICE, even in shallow waters, confirming that this region, unlike all others around Gondwana, retained a shallow redoxcline and experienced no increase in oxygenation post-SPICE. We conclude that under the generally low-atmospheric-oxygen state of the Cambrian, dysoxia was prevalent in some regions even in very shallow waters, with the apparent global variability in shallow-water redox conditions likely reflecting local productivity controls and/or upwelling dynamics.

ACKNOWLEDGEMENTS

This work was supported by UK Research and Innovation (UKRI) projects NE/T008458/1, NE/Z000122, and EP/Y008790/1. We thank M. Yilales Agelvis, S. Reid, Y. Xiong, and A. Hobson for technical support, and NatureScot for permission to sample the Eilean Dubh Formation at Durness. We thank the anonymous reviewers of this work.

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