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Science Paper

Carbon Isotope Trends of Precambrian-Cambrian Carbonates in Southwestern Laurentia Are Robust to Diagenetic Overprinting

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An extreme negative (<-6‰) carbon isotope ($\delta^{13}\text{C}$) excursion is recognized globally in strata recording the Ediacaran-Cambrian boundary. This excursion has been termed the BASal Cambrian carbon isotope Excursion (BACE). It, like other carbon isotope excursions throughout the geologic record, has been interpreted to record a perturbation to the global carbon cycle based on the assumption that shallow water carbonate sediments and rock preserve an accurate time-series of the $\delta^{13}\text{C}$ composition of the global ocean. However, this assumption has been demonstrated to be inaccurate in some shallow water settings; pervasive early diagenetic alteration of shallow water carbonate $\delta^{13}\text{C}$ records and local carbon cycling within the platform environment can result in the decoupling of global ocean and shallow water carbonate $\delta^{13}\text{C}$ values.

Here, we test the extent and isotopic effects of early diagenetic alteration of shallow water carbonate sediments that record the BACE in southwestern Laurentia using carbonate stable isotope ($\delta^{13}\text{C}$, $\delta^{18}\text{O}$, $\delta^{44/40}\text{Ca}$, $\delta^{26}\text{Mg}$) and major and minor element (Mg/Ca, Sr/Ca) geochemistry. The $\delta^{44/40}\text{Ca}$ values of the three studied sections are consistent with different modes of diagenetic alteration, ranging from more sediment-buffered to more fluid-buffered early marine diagenesis. Dolomite $\delta^{26}\text{Mg}$ and $\delta^{18}\text{O}$ values suggest that diagenetic alteration and dolomitization occurred prior to significant burial. Because the $\delta^{13}\text{C}$ chemostratigraphic records are reproducible despite the variable diagenetic regimes among the three sites, we argue that the prominent $\delta^{13}\text{C}$ stratigraphic trends were insensitive to diagenetic overprinting and instead reflect changes in the primary $\delta^{13}\text{C}$ composition of dissolved inorganic carbon of platform seawater. Finally, we explore a potential link between the BACE, an increase in early marine fluid-buffered marine diagenesis of platform carbonates, and magmatism associated with the rifting of the southwest margin of Rodinia.

1. INTRODUCTION

Interpretations of global carbon isotope ($\delta^{13}\text{C}$) excursions in shallow water (<200 m water depth) carbonate sediments and rocks inform models of global environmental change across Earth history, often over intervals of biotic turnover. Classically, these interpretations assume platform carbonate $\delta^{13}\text{C}$ values approximate the $\delta^{13}\text{C}$ composition of dissolved inorganic carbon (DIC) within the global ocean (Kump & Arthur, 1999). This assumption is not testable prior to the proliferation of planktic calcifiers in the Mesozoic and consequent expansion of the carbonate archive beyond the continental shelf. However, in the Cenozoic, this assumption has been shown to sometimes be invalid. For

example, records of $\delta^{13}\text{C}$ values in shallow water carbonate sediments over the last 10 million years exhibit $\delta^{13}\text{C}$ values that are several per mil more positive than the $\delta^{13}\text{C}$ values from the open ocean over the same time interval (Swart, 2008; Swart & Eberli, 2005), providing empirical evidence that shallow water carbonate sediments across the globe can be systematically, synchronously decoupled from open-ocean inorganic carbon pools. Even during perturbations to the global carbon cycle caused by the addition of large amounts of carbon depleted in ^{13}C (relative to atmospheric CO_2), like that interpreted at the Paleocene–Eocene Thermal Maximum (PETM), shallow marine carbonates preserve $\delta^{13}\text{C}$ trends quantitatively distinct from those recorded in deep and surface ocean archives (Li et al., 2020, 2021).

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Shallow water carbonate sediments form in environments that are not as well connected to the large reservoirs of inorganic carbon in the ocean-atmosphere system as deeper water carbonate facies. Compared to pelagic carbonates, $\delta^{13}\text{C}$ values of shallow water carbonate sediments are characterized by a much larger range of values compared to pelagic carbonates, which are controlled by a wide range of carbonate sources (e.g., microbes, algae, invertebrates, abiogenic particles) and local processes (e.g., Geyman & Maloof, 2019). Local phenomena that affect the $\delta^{13}\text{C}$ composition of shallow water carbonate sediment can have global drivers; for example, changes in eustatic sea-level, resulting in an expansion of platform areas, have been hypothesized to generate synchronous $\delta^{13}\text{C}$ excursions in platform sediments during the late Ediacaran Period through enhanced productivity and/or evaporation in shallow marine environments (Busch et al., 2022). Additionally, studies of shallow water marine carbonate sediments indicate that early diagenetic alteration and associated changes in mineralogy can alter the chemical composition of the carbonate sedimentary archive after deposition (Fantle & Higgins, 2014; Higgins et al., 2018; Swart & Eberli, 2005). In particular, carbonate $\delta^{44/40}\text{Ca}$, $\delta^{26}\text{Mg}$, and Sr/Ca records have been shown to systematically covary during early diagenesis (Ahm et al., 2019; Blättler et al., 2015; Higgins et al., 2018) and have been used to identify local alteration of shallow marine carbonate $\delta^{13}\text{C}$ records of ancient carbonate strata (e.g., Ahm et al., 2021; D. S. Jones et al., 2020). Thus, while global $\delta^{13}\text{C}$ excursions preserved in shallow marine carbonate $\delta^{13}\text{C}$ records can reflect global perturbations to the carbon cycle (e.g., during the PETM), they can also (or instead) record local forcings (which may, in some cases, respond to global sea level).

A global $<-6\text{‰}$ $\delta^{13}\text{C}$ excursion is documented from shallow water carbonate $\delta^{13}\text{C}$ records that span the Ediacaran–Cambrian (539–533 Ma) transition (e.g., Derry et al., 1992; Kaufman & Knoll, 1995; Magaritz et al., 1991; L. L. Nelson et al., 2023), a critical interval in the evolutionary history of life that saw turnover between early animal communities prior to the major radiation of motile and biomineralizing animals in the Cambrian Period. The excursion, termed the BASal Cambrian carbon isotope Excursion (BACE) (Zhu et al., 2006), is considered to be broadly correlative among Ediacaran–Cambrian boundary sections because, in western Laurentia, it directly underlies the first occurrence of the ichnofossil *Treptichnus pedum*, the marker of the Cambrian Global Boundary Stratotype Section and Point (GSSP) (Brasier et al., 1994; Corsetti & Hagadorn, 2000; Narbonne et al., 1994). Interpreted as a record of global carbon cycle perturbation, the BACE has been used as evidence for environmental change immediately preceding the Cambrian Period with potential mechanistic links to end-Ediacaran biotic turnover (e.g., Amthor et al., 2003; Hodgkin et al., 2021; Kimura et al., 1997; Kimura & Watanabe, 2001; Smith et al., 2023). It has also been used to identify the Ediacaran–Cambrian boundary where *T. pedum* does not appear due to taphonomic bias (e.g., Maloof et al., 2005; Zhou & Xiao, 2007), resulting in the development of regional and global chemostratigraphic age models

for the Ediacaran–Cambrian transition (e.g., Bowyer et al., 2022; Maloof et al., 2010; L. L. Nelson et al., 2023; Smith et al., 2016).

However, there are reasons to suspect the BACE may not record a global carbon cycle perturbation. Due to the lack of a precise geochronological framework that calibrates the chemostratigraphic record across the Ediacaran–Cambrian boundary and the fact that there are several large negative carbon isotope excursions in the latest Ediacaran and earliest Cambrian, the BACE is not independently confirmed to be globally synchronous. This has manifested in several different stratigraphic age models for this interval of Earth history (e.g., Bowyer et al., 2022, 2024; L. L. Nelson et al., 2023). Additionally, it is difficult to explain the generation of global ocean DIC $\delta^{13}\text{C}$ values $<-6\text{‰}$, especially for $>10^5$ – 10^6 years, based on modern steady-state carbon cycling. The $\delta^{13}\text{C}$ composition of carbon entering the ocean reservoir is assumed to approximate mantle values of $\sim-6\text{‰}$ (Kump & Arthur, 1999), which, over long time-scales, should equal the cumulative $\delta^{13}\text{C}$ composition of carbon exiting the system via organic and inorganic carbon burial. However, this assumption of mantle value inputs does not always hold true. For example, in the Neogene, it has been shown that recycling of sedimentary kerogen and geogenic methane can drive the mean $\delta^{13}\text{C}$ input to $\sim-8\text{‰}$ (e.g., Derry, 2024). Photosynthesis fractionates carbon isotopes such that organic carbon is depleted in ^{13}C relative to the mantle, requiring carbonate $\delta^{13}\text{C}$ values to be greater than or, if no organic burial occurs, equal to mantle values. Shifts in carbon weathering inputs and in fractionation during photosynthesis can also impact $\delta^{13}\text{C}$ records (Kump et al., 1999; Vogel, 1993), but neither of these have been proposed as viable drivers of a negative carbon isotope excursion of similar magnitude as the BACE. To generate a global carbonate $\delta^{13}\text{C}$ excursion $<-6\text{‰}$, a large external pool of isotopically light carbon is needed; the $\delta^{13}\text{C}$ composition of ocean DIC may reach values $<-6\text{‰}$ only if (1) a large amount of light carbon enters the ocean-atmosphere system or (2) a large amount of heavy carbon is removed from it. Because the BACE reaches below modern mantle $\delta^{13}\text{C}$ values, models proposed to explain it as a record of global average DIC must invoke unconventional external carbon sources (e.g., Bjerrum & Canfield, 2011; Hodgkin et al., 2021; Rothman et al., 2003; Svensen et al., 2004) or sinks (e.g., Laakso & Schrag, 2020).

Given these complications and those associated with interpreting the shallow water carbonate archive, it is reasonable to consider whether hypotheses that do not require a global carbon cycle perturbation can explain both the geochemical character and global nature of the BACE. Here, we revisit three carbonate-rich successions from the western margin of Laurentia, where the relationship between the BACE and the Ediacaran–Cambrian boundary was first established. We characterize the role of early diagenetic alteration in setting the chemical composition of carbonate sediments across the BACE using a suite of stable isotope ($\delta^{13}\text{C}$, $\delta^{18}\text{O}$, $\delta^{44/40}\text{Ca}$, $\delta^{26}\text{Mg}$) and trace elements (Sr/Ca) on carbonate records in concert with sedimentary observations. We find evidence that shallow water carbonate sed-

imentary strata recording the BACE are persistently influenced by early diagenetic alteration, yet based on geochemical fingerprints, the extent and nature of interpreted diagenetic regimes differ among the three study sites. Despite stratigraphic and lateral variability in the diagenetic alteration of preserved carbonates, the stratigraphic pattern of $\delta^{13}\text{C}$ values across the BACE are consistent among the three sites. We discuss these results in the context of possible origins of the BACE and interpretations of carbon isotope excursions in shallow water carbonate sediments.

2. GEOLOGIC BACKGROUND

Ediacaran–Cambrian boundary strata of southwestern Laurentia record mixed siliciclastic-carbonate sedimentation during the protracted break-up of Rodinia (Armin & Mayer, 1983; Bond & Kominz, 1984; Fedo & Cooper, 2001; Karlstrom et al., 2020). These strata range from fluvio-marine deltaic to outer-shelf deposits, deepening to the west-northwest in present-day coordinates (Fedo & Cooper, 2001; C. A. Nelson, 1978; Smith et al., 2023; Stewart, 1970). The Ediacaran–Cambrian boundary, marked by the first occurrence of *T. pedum*, is recorded in correlative units from three regions: the Esmeralda Member of the Deep Spring Formation of the White and Inyo Ranges and Esmeralda County (CA and NV, USA), the lower member of the Wood Canyon Formation of the Death Valley region (CA and NV, USA), and the La Ciénega and overlying Cerro Rajón formations of the Caborca region (Mexico; [fig. 1](#)) (Corsetti & Hagadorn, 2003; Hagadorn & Waggoner, 2000; Loyd et al., 2012).

Litho- and sequence stratigraphy form the backbone for the correlation of Ediacaran–Cambrian strata from the southwestern United States (Stewart, 1970; Wheeler, 1948). Over the past several decades, bio- and chemostratigraphy have been used to substantiate, refine, and extend these correlations to outcrops near Caborca, Mexico (Corsetti & Hagadorn, 2000; Hodgkin et al., 2021; Jensen et al., 2002; Loyd et al., 2012; Oliver & Rowland, 2002; Sour-Tovar et al., 2007; Stewart et al., 1984). Late Ediacaran body fossils are reported from Unit 1 of the La Ciénega Formation (Hodgkin et al., 2021; Schiffbauer et al., 2024; Sour-Tovar et al., 2007; Stewart et al., 1984), the Dunfee Member of the Deep Spring Formation (Grant, 1990; Mount et al., 1983; Rivas et al., 2024; Selly et al., 2020; Signor et al., 1987; Smith et al., 2016, 2023; Taylor, 1966), and the first parasequence in the lower member of the Wood Canyon Formation (Evans et al., 2024; Hagadorn & Waggoner, 2000; Hall et al., 2020; Selly et al., 2020; Smith et al., 2017). Although moderately complex trace fossils have been found in upper Ediacaran strata at Mount Dunfee (Tarhan et al., 2020), the lowest occurrence of *T. pedum* is in the top ~75 m of the Esmeralda Member of the Deep Spring Formation (Corsetti & Hagadorn, 2003), at the base of the Cerro Rajón Formation (Barrón-Díaz et al., 2019; Hodgkin et al., 2021; Loyd et al., 2012), and meters above the second dolostone interval of the lower Wood Canyon Formation (Corsetti & Hagadorn, 2000; Hagadorn & Waggoner, 2000).

The BACE is preserved across southwestern Laurentia, spanning sequence boundaries and variable carbonate facies, textures, and lithologies (Smith et al., 2023). It was first identified in southwestern Laurentia within the Dunfee and Esmeralda members of the Deep Spring Formation (Corsetti & Kaufman, 1994). There, a $\delta^{13}\text{C}$ excursion with a nadir of -9.5‰ is preserved in limestone and dolostone strata (Corsetti & Kaufman, 1994; Smith et al., 2016, 2023; [fig. 1B](#)). A correlative $\delta^{13}\text{C}$ excursion was later identified in the more proximal to paleo-shoreline Wood Canyon Formation, spread across the three dolostone units of the lower member (Corsetti & Hagadorn, 2000; [fig. 1B](#)). Although thick siliciclastic packages separate the dolostone units, obscuring the full expression of the BACE, a nadir of $\sim -9\text{‰}$ is observed in the first dolostone unit (Corsetti & Hagadorn, 2000; Smith et al., 2023). In the lower member of the Wood Canyon Formation, a chemical abrasion isotope-dilution thermal ionization mass spectrometry (CA-ID-TIMS) U-Pb detrital zircon maximum depositional age of 532.83 ± 0.98 Ma coincides with the first positive $\delta^{13}\text{C}$ excursion above the BACE (L. L. Nelson et al., 2023). Strata from the Wood Canyon and Deep Spring formations are also chemostratigraphically correlated with units 2 and 3 of the La Ciénega Formation, in which the BACE reaches $\delta^{13}\text{C}$ values as low as -7.5‰ over ~50 m of dolostones and interbedded siliciclastic rocks (Hodgkin et al., 2021; Loyd et al., 2012; [fig. 1B](#)). In the La Ciénega Formation, a CA-ID-TIMS U-Pb detrital zircon maximum depositional age of 539.40 ± 0.23 Ma was obtained from strata deposited ~20 m above the nadir of the BACE and ~10 m below the return to -0‰ $\delta^{13}\text{C}$ values (Hodgkin et al., 2021). This maximum depositional age constrains the ascending limb of the BACE in Laurentia to ≤ 540 Ma. Previously, the southwestern Laurentian record of the BACE has been argued to be robust to alteration via meteoric diagenesis based on a lack of a clear relationship between $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values of the BACE and the fact that the excursion stratigraphically spans multiple sequence boundaries and is laterally extensive over several hundreds of kilometers (L. L. Nelson et al., 2023; Smith et al., 2016, 2023).

3. MATERIALS AND METHODS

Samples were collected from three sites within the southwestern Great Basin ([fig. 1A](#)): Mount Dunfee, Nevada, USA (37.34162° , -117.31937°); Echo Canyon, California, USA (36.51906° , -116.66620°); and Cerro Rajón, Sonora, Mexico (30.41727° , -111.94530°). New geochemical data, including carbonate calcium isotope (‰ , $n=173$) and magnesium isotope (‰ , $n=42$) compositions and Mg/Ca (mmol/mol), Sr/Ca (mmol/mol), Mn/Ca ($\mu\text{mol/mol}$), U/Ca ($\mu\text{mol/mol}$) major and trace elemental ratios ($n=186$), are paired with previously published sedimentological observations, $\delta^{13}\text{C}$ values, and $\delta^{18}\text{O}$ values from the top 45 m of the Dunfee Member of the Deep Spring Formation and the entirety of the Esmeralda Member of the Deep Spring Formation at Mount Dunfee (Smith et al., 2016), the lower member of the Wood Canyon Formation at Echo Canyon (Smith et al., 2023), and the La Ciénega Formation at Cerro Rajón (Hodgkin et al.,

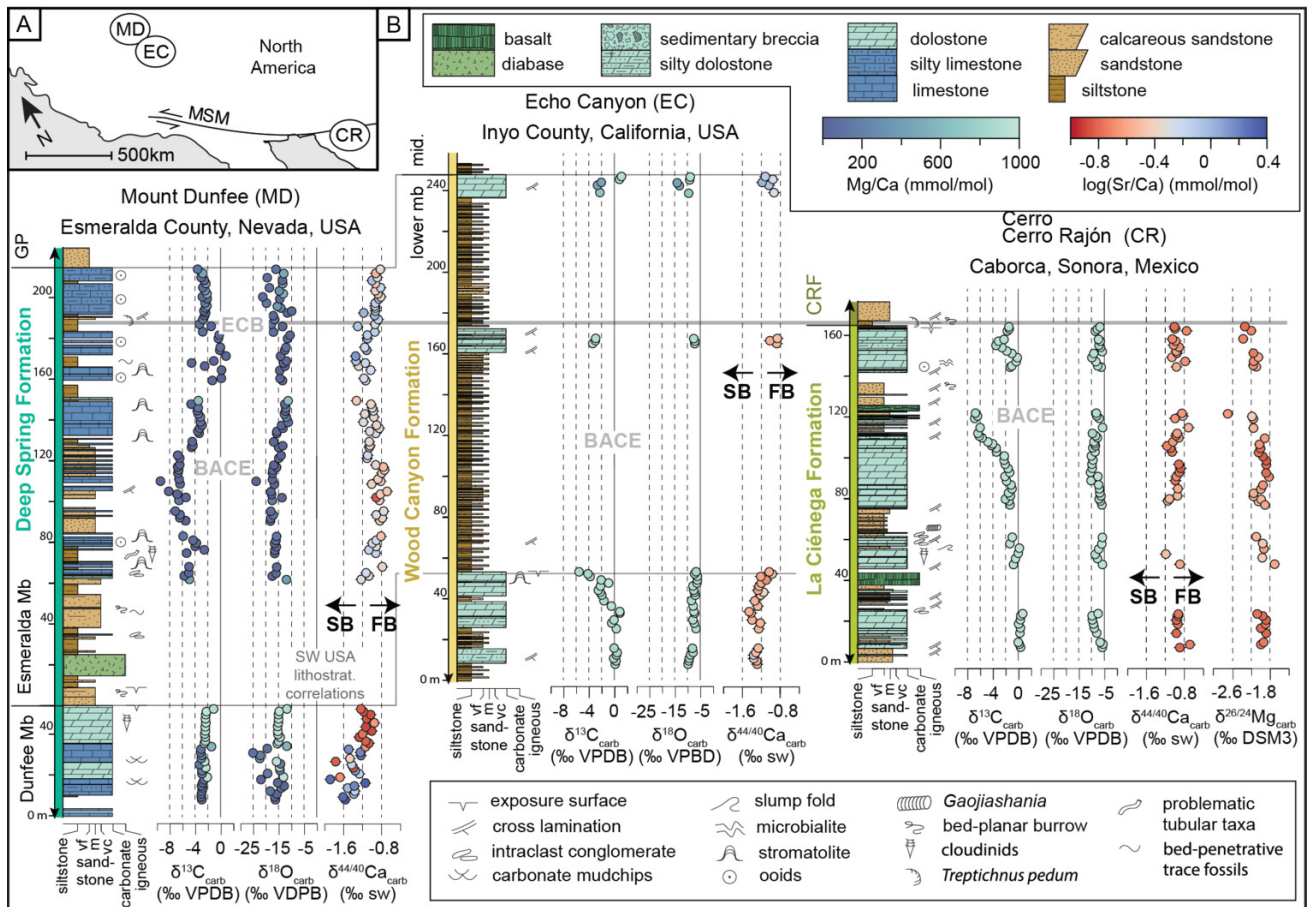


Figure 1. (A) Locality map for study sites. Study sites include (1) Mount Dunfee in Esmeralda County, NV, USA, (2) Echo Canyon in Death Valley, CA, USA, and (3) Cerro Rajón in Sonora, Mexico. (B) Litho-, bio-, and chemo-stratigraphy of the Deep Spring, Wood Canyon, and La Ciénega formations. For the La Ciénega, Wood Canyon, and Deep Spring formations, $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values are plotted against stratigraphic thickness and are color-scaled by Mg/Ca ratios. The $\delta^{44/40}\text{Ca}$ values are plotted against stratigraphic thickness and are colored-scaled by Sr/Ca ratios (log-scaled). For the La Ciénega Formation, $\delta^{26/24}\text{Mg}$ values are plotted against stratigraphic thickness and colored by Sr/Ca ratios. Lithostratigraphic, $\delta^{13}\text{C}$, and $\delta^{18}\text{O}$ data from Mount Dunfee, Cerro Rajón, and Echo Canyon are from Smith et al. (2016), Hodgin et al. (2021), and Smith et al. (2023), respectively. BACE=Basal Cambrian carbon isotope Excursion. ECB=Ediacaran-Cambrian Boundary; SB=sediment-buffered; FB=fluid-buffered.

2021). All geochemical data were generated from the same carbonate powders, originally produced by Hodgin et al. (2021), Smith et al. (2016), and Smith et al. (2023) by micro-drilling cut carbonate samples, with care taken to avoid veins, fractures, and siliciclastic-rich laminae.

Facies associations are defined for the Esmeralda and upper Dunfee members of the Deep Spring Formation (after Wilson, 1974). They are based on field observations from Smith et al. (2016) and on new observations that were made as part of this study while retracing the stratigraphic section of Smith et al. (2016). High order transgressive-regressive cycles (after Catuneanu, 2017; Embry, 2002) were then identified in the Esmeralda Member, informed by stratigraphic changes in lithologies and carbonate facies. Additionally, twenty-three petrographic thin sections of carbonates from the Deep Spring Formation at Mount Dunfee were made from the samples that sourced powders. Samples were selected for thin-section based on their (1) cal-

cium isotope composition and (2) facies assignment, with the goal of creating a sample set that reflected the variation in calcium isotope values and facies recorded throughout the stratigraphic section. Through petrographic observation of thin-sections, sample textures were characterized.

3.1. Trace and major element analyses

Following dissolution in buffered acetic acid (pH ~5) at Johns Hopkins University, samples were analyzed for elemental ratios at Princeton University using a Thermo Finnegan iCAP Q Inductively Coupled Plasma Mass Spectrometer (ICP-MS), measured against a set of in-house (matrix-matched modern seawater and aragonite) and NIST standards (SRM88b). The external reproducibility of Sr/Ca and Mg/Ca are estimated from measurements of SRM88b to be <5% (2 σ , n=16). The external reproducibilities of U/Ca and Mn/Ca ratios are estimated from measurements of SRM88b to be <12% and <3%, respectively (2 σ , n=16).

3.2. $\delta^{44/40}\text{Ca}$ and $\delta^{26}\text{Mg}$ analyses

Samples were prepared and analyzed for calcium and magnesium isotope analyses at Princeton University. They were processed using an automated high-pressure ion chromatography (IC) system (Dionex ICS-5000+) and run on a Thermo Scientific Neptune Plus multi-collector ICP-MS following methods outlined in detail in Husson et al. (2015). The analyses were conducted in medium resolution mode, measuring ^{44}Ca , ^{43}Ca , and ^{42}Ca using the sample-standard bracketing method. Calcium isotopic compositions are reported in delta notation as the relative abundances of ^{44}Ca and ^{40}Ca normalized against a modern seawater standard, assuming that the kinetic isotope mass fractionation law governs the relationship between $\delta^{44/40}\text{Ca}$ and $\delta^{44/42}\text{Ca}$ (e.g., Gussone et al., 2020). The external reproducibility of $\delta^{44/40}\text{Ca}$ measurements of SRM915b is $-1.18 \pm 0.17\text{‰}$ (2σ , $n=44$). Magnesium isotopic compositions are reported in delta notation as the relative abundances of ^{26}Mg and ^{24}Mg normalized against Dead Sea Magnesium (DSM3). The external reproducibility of $\delta^{26}\text{Mg}$ measurements of Cambridge-1 and modern seawater standards are $-2.60 \pm 0.09\text{‰}$ (2σ , $n=8$) and $-0.82 \pm 0.10\text{‰}$ (2σ , $n=8$), respectively.

4. RESULTS

4.1. The Deep Spring Formation at Mount Dunfee (Esmeralda County, NV)

The measured section of the Deep Spring Formation at Mount Dunfee records stratigraphic variation in $\delta^{13}\text{C}$, $\delta^{18}\text{O}$, $\delta^{44/40}\text{Ca}$, and Sr/Ca values in both the Dunfee and Esmeralda members. In the uppermost 45 m of the Dunfee Member, there are interfingering limestone, dolomitic limestone, and dolostone intervals. Transitions from limestone to dolostone occur at ~20 m and ~40 m below the top of the member, but there is variability in these lithologies along strike (fig. 1B). These mineralogical shifts were observed in the field and confirmed using Mg/Ca mmol/mol ratios, with the cutoff between limestone to dolostone defined here at 600 mmol/mol. The transition to dolostone coincides with the stabilization of $\delta^{18}\text{O}$ values averaging ~-15‰ (from values down to -25‰ recorded in the limestone), a shift in $\delta^{13}\text{C}$ values from ~-3‰ to ~-2‰, and a decrease in Sr/Ca ratios (figs. 1B and 2). This transition towards the top of the Dunfee Member also coincides with a shift in $\delta^{44/40}\text{Ca}$ values, in which dolostones in the upper 20 m of the member record $\delta^{44/40}\text{Ca}$ values between -1.27‰ and -0.95‰, and the underlying limestones and dolostones record $\delta^{44/40}\text{Ca}$ between -1.89‰ and -1.24‰. The Sr/Ca values of the limestone samples in the Dunfee Member are the highest of any of the samples measured but are low when compared to Sr/Ca values from the Bahamas platform (fig. 3).

In the limestone-dominated Esmeralda Member, $\delta^{44/40}\text{Ca}$ values range from -1.3‰ to -0.7‰, while Sr/Ca ratios range from 0.1 mmol/mol to 1.2 mmol/mol. Stratigraphically, $\delta^{44/40}\text{Ca}$ values show a systematic trend reaching highs (~0.8‰) at ~100 m and ~200 m above the base of

the member (fig. 1B). Esmeralda limestones with high ($\geq -0.9\text{‰}$) $\delta^{44/40}\text{Ca}$ values have Sr/Ca ratios between 0.09 mmol/mol and 0.93 mmol/mol, while those with lower $\delta^{44/40}\text{Ca}$ values have Sr/Ca ratios between 0.26 mmol/mol and 1.23 mmol/mol (fig. 2B). Relative to Sr/Ca values measured from sediment from sites on and around the Bahamas platform, all the values measured in this study, even those with more negative or sediment-buffered $\delta^{44/40}\text{Ca}$ values, are relatively low (fig. 3). Covariation is also observed between $\delta^{44/40}\text{Ca}$, $\delta^{18}\text{O}$, and $\delta^{13}\text{C}$ values in the Esmeralda Member. This covariation is most evident over the stratigraphic interval that records the ascending limb of the BACE, where $\delta^{44/40}\text{Ca}$ values decrease from ~-0.7 to -1.3‰, $\delta^{18}\text{O}$ values increase from ~-18 to -9‰, and $\delta^{13}\text{C}$ values increase from values as low as ~-9 to 1‰ (figs. 1B, 2A, and 4).

Of the three sections studied, the stratigraphic section of the Deep Spring Formation is the thickest and best texturally preserved. As such, it records the greatest diversity in carbonate facies. In the field, no macroscopic evidence of authigenic carbonate precipitation (e.g., calcite nodules, infilled void spaces) was observed. Seven carbonate facies associations were defined within this unit and paired with $\delta^{44/40}\text{Ca}$ and $\delta^{13}\text{C}$ data (FA1-FA7, table 1; fig. 5). FA1 and FA2 are interpreted to record outer shelf sedimentation just below and at fair-weather wave base, respectively. These facies are differentiated by the abundance of siltstone partings in FA1 and the preservation of low-angle cross-lamination in wackestones from FA2. FA3 consists of oolites, interpreted to record high-energy ooid shoals positioned on the outer to mid-shelf. Together, FA1, FA2, and FA3 comprise much of the Esmeralda Member. FA4 consists of stromatolites and limestone wackestone-grainstones, interpreted to record carbonate deposition in shelf environments shielded from high-energy conditions. The shallowest carbonate facies preserved in the Esmeralda Member are assigned to FA5, which consists of sandy limestone grainstone lenses among fine quartz-rich sandstones. FA5 is interpreted to record inner shelf sedimentation. Strata with FA5 facies assignment record the highest $\delta^{44/40}\text{Ca}$ and lowest $\delta^{13}\text{C}$ values (fig. 5). Strata belonging to FA6 and FA7 are reported only from the Dunfee Member. FA6 is defined by lime mudstone deposits with an abundance of micritic chip horizons, from which an inner shelf depositional environment is interpreted, while FA7 consists of fabric-destructive dolostones associated with a karstic dissolution surface, observed at the top of the Dunfee Member (fig. 1B).

In thin-section, the original depositional fabrics of carbonates are poorly preserved regardless of facies association, although relics of original structures remain in some samples (e.g., FA3-c in fig. 5). No correlation is observed between fabric retention and $\delta^{44/40}\text{Ca}$ and $\delta^{13}\text{C}$ trends recorded in limestone strata. For example, samples FA3-a and FA3-b record similar $\delta^{44/40}\text{Ca}$ and $\delta^{13}\text{C}$ values, but the fabric of FA3-b is more thoroughly replaced by neomorphic spar (fig. 5). Additionally, within FA5, the relative proportion of quartz grains does not clearly relate to $\delta^{44/40}\text{Ca}$ and $\delta^{13}\text{C}$ values; among the six FA5 samples observed in thin-section and 16 observed in hand sample, no trend was ob-

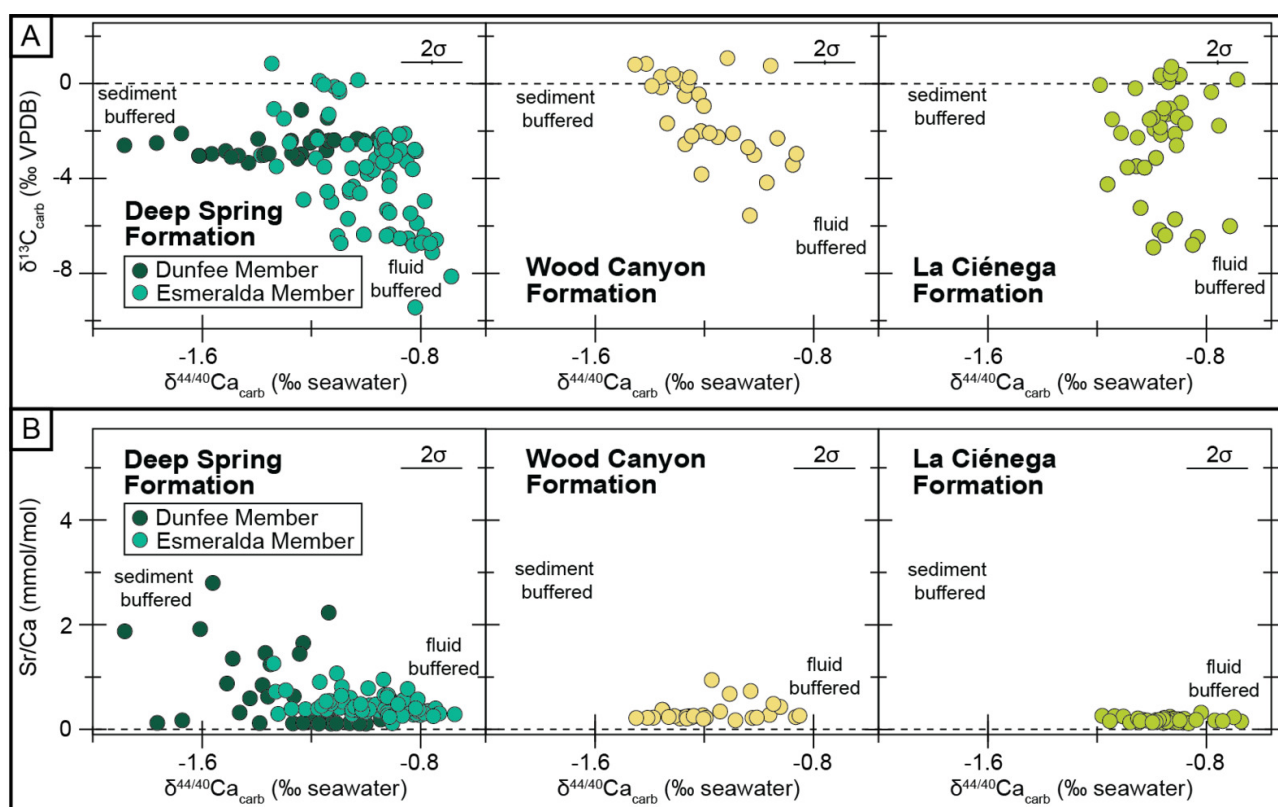


Figure 2. (A) Cross-plots of $\delta^{44/40}\text{Ca}$ and $\delta^{13}\text{C}$ records from the Deep Spring, Wood Canyon, and La Ciénega formations. A negative relationship between $\delta^{44/40}\text{Ca}$ and $\delta^{13}\text{C}$ values is observed in the Esmeralda Member of the Deep Spring Formation and in the Wood Canyon Formation. The $\delta^{44/40}\text{Ca}$ and $\delta^{13}\text{C}$ of the Dunfee Member show a different trend from those of the Esmeralda Member. All these values are below a karstic exposure surface and within a stratigraphic interval that is partially dolomitized (see [fig. 1B](#)). The $\delta^{13}\text{C}$ data from Mount Dunfee, Cerro Rajón, and Echo Canyon are from Smith et al. (2016), Hodgins et al. (2021), and Smith et al. (2023), respectively. (B) Cross-plot of $\delta^{44/40}\text{Ca}$ and Sr/Ca records from the Deep Spring, Wood Canyon, and La Ciénega formations. The relationship between $\delta^{44/40}\text{Ca}$ values and Sr/Ca ratios observed in the Deep Spring Formation matches the marine diagenesis model results of Ahm et al. (2018).

served between quartz content and $\delta^{44/40}\text{Ca}$ or $\delta^{13}\text{C}$ values ([fig. 5](#)).

4.2. The Wood Canyon Formation at Echo Canyon (Death Valley region)

The samples from the lower Wood Canyon Formation at Echo Canyon are dolostone and record a shift in $\delta^{44/40}\text{Ca}$ values from $\sim -1.4\text{‰}$ to $\sim -1.0\text{‰}$. This $\sim -0.4\text{‰}$ shift is coincident with the downturn of the BACE and occurs in the basal 55 m of the section. Following an interval dominated by siliciclastic strata, carbonates re-appear at 158 m to 161 m, and 232 m to 240 m into the section, and record $\delta^{44/40}\text{Ca}$ values between -0.9‰ and -1.2‰ . The Sr/Ca ratios are consistently low, with values between 0.15 and 0.92 mmol/mol (figs. [1B](#) and [2B](#)).

4.3. The La Ciénega Formation at Cerro Rajón (Caborca region)

Carbonate strata from the section of the La Ciénega Formation at Cerro Rajón consist of dolostone with Sr/Ca ratios < 0.3 mmol/mol (figs. [1B](#) and [2B](#)). These dolostones record $\delta^{44/40}\text{Ca}$ values that oscillate between -0.7‰ and -1.2‰

over ~ 5 m to ~ 20 m-thick intervals. Strata that record the descending limb of the BACE record a $\sim -0.5\text{‰}$ shift in $\delta^{44/40}\text{Ca}$ values from -1.2‰ to 0.7‰ , but $\delta^{44/40}\text{Ca}$ and $\delta^{13}\text{C}$ values generally do not covary in this section (figs. [1B](#) and [2](#)). The La Ciénega Formation also records stratigraphic fluctuation in $\delta^{26}\text{Mg}$ values. Over the lower 122 m of the section, $\delta^{26}\text{Mg}$ values oscillate between -2.7‰ and -1.7‰ . Over the upper 20 m of the section, $\delta^{26}\text{Mg}$ values decrease from -2.0‰ to -2.3‰ . No consistent relationship is observed between $\delta^{44/40}\text{Ca}$ and $\delta^{26}\text{Mg}$ trends ([fig. 1B](#)). Metabasalt sills intrude carbonates that record the BACE at the Cerro Rajón site (Tapia-Trinidad et al., 2022), raising the potential for carbonate interaction with hydrothermal fluids during basalt emplacement.

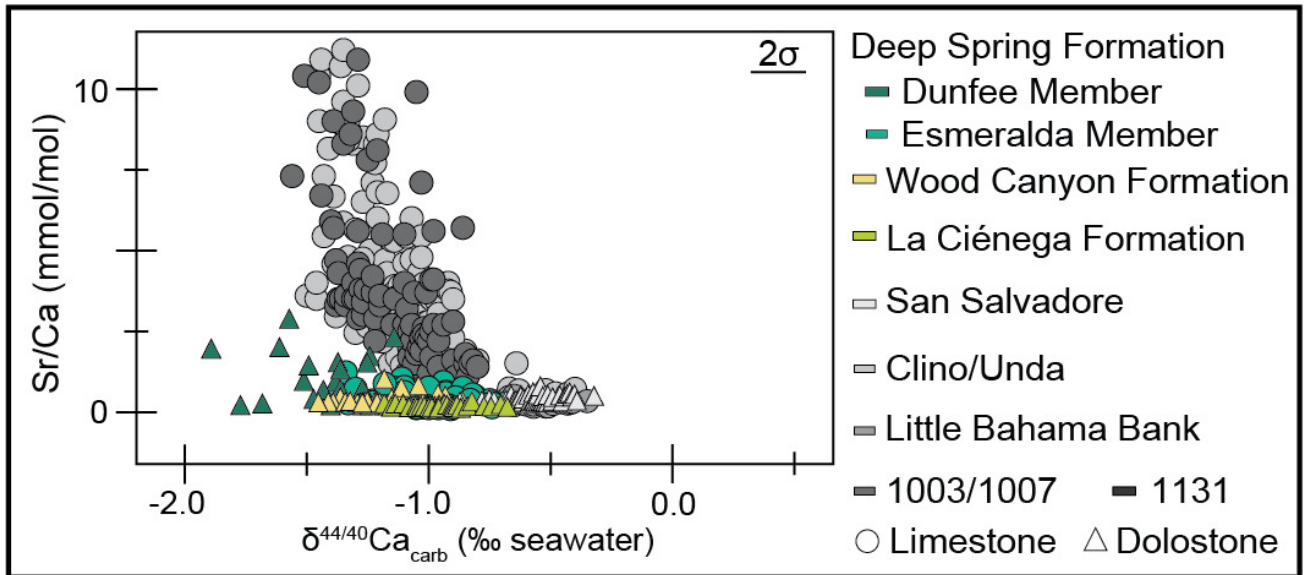


Figure 3. Cross-plot of $\delta^{44/40}\text{Ca}$ and Sr/Ca values from the Deep Spring, Wood Canyon, and La Ciénega formations (this study) and from sediments from sites on and adjacent to the Bahamas platform (Ahm et al., 2018; Higgins et al., 2018).

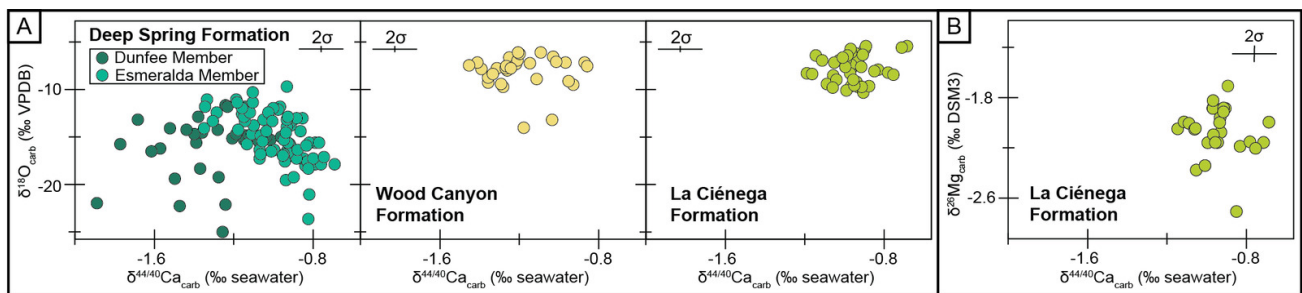


Figure 4. (A) Cross-plots of $\delta^{18}\text{O}$ and $\delta^{44/40}\text{Ca}$ records from the Deep Spring Formation, Wood Canyon Formation, and La Ciénega Formation. Outlier points, which record low $\delta^{18}\text{O}$ and $\delta^{44/40}\text{Ca}$ values, are from the Dunfee Member of the Deep Spring Formation. The $\delta^{18}\text{O}$ data from Mount Dunfee, Cerro Rajón, and Echo Canyon are from Smith et al. (2016), Hodgins et al. (2021), and Smith et al. (2023), respectively. (B) Cross-plot of $\delta^{44/40}\text{Ca}$ and $\delta^{26}\text{Mg}$ records from the La Ciénega Formation.

5. DISCUSSION

5.1. Controls on $\delta^{44/40}\text{Ca}$ trends in southwestern Laurentia

5.1.1. Influence of global calcium cycling

Stratigraphic trends in $\delta^{44/40}\text{Ca}$ records across southwestern Laurentia are unlikely to be caused by temporal changes in the $\delta^{44/40}\text{Ca}$ value of the ocean. Since the main calcium sink from the ocean is carbonate burial, bulk carbonate $\delta^{44/40}\text{Ca}$ values cannot change over long timescales when the calcium cycle must be in steady state; or, put differently, the $\delta^{44/40}\text{Ca}$ value of outputs must match those of the inputs for timescales >1 Myr (Gussone et al., 2020; Silva-Tamayo et al., 2018). Seawater $\delta^{44/40}\text{Ca}$ values are also well-buffered against transient (<1 Myr) changes caused by temporary imbalances in calcium sources and sinks. This stability is controlled by carbonate compensation, in which

changes in calcium input or outputs to the ocean are rapidly buffered by the carbonate saturation state (Ω). For example, an increase in calcium sources would cause an increase in the saturation state, leading to precipitation of carbonate minerals. Modeling studies of the combined calcium and carbon cycles incorporating this effect have demonstrated that flux perturbations can only result in a maximum $\delta^{44/40}\text{Ca}$ excursion of $<0.3\text{‰}$ (Komar & Zeebe, 2011, 2016).

Seawater $\delta^{44/40}\text{Ca}$ values can only shift if there is a significant change in the fractionation between the bulk carbonate sink and seawater. For example, a shift in the dominant mode of carbonate precipitation from aragonite to calcite could cause a change in the $\delta^{44/40}\text{Ca}$ value of seawater of $<0.5\text{‰}$. However, for this type of global perturbation to show up in the bulk carbonate record, it would have to occur rapidly, over <1 Myr time-scales, before the calcium cycle reaches a new steady-state. Previous calcium isotope studies of the Ediacaran–Cambrian transition show

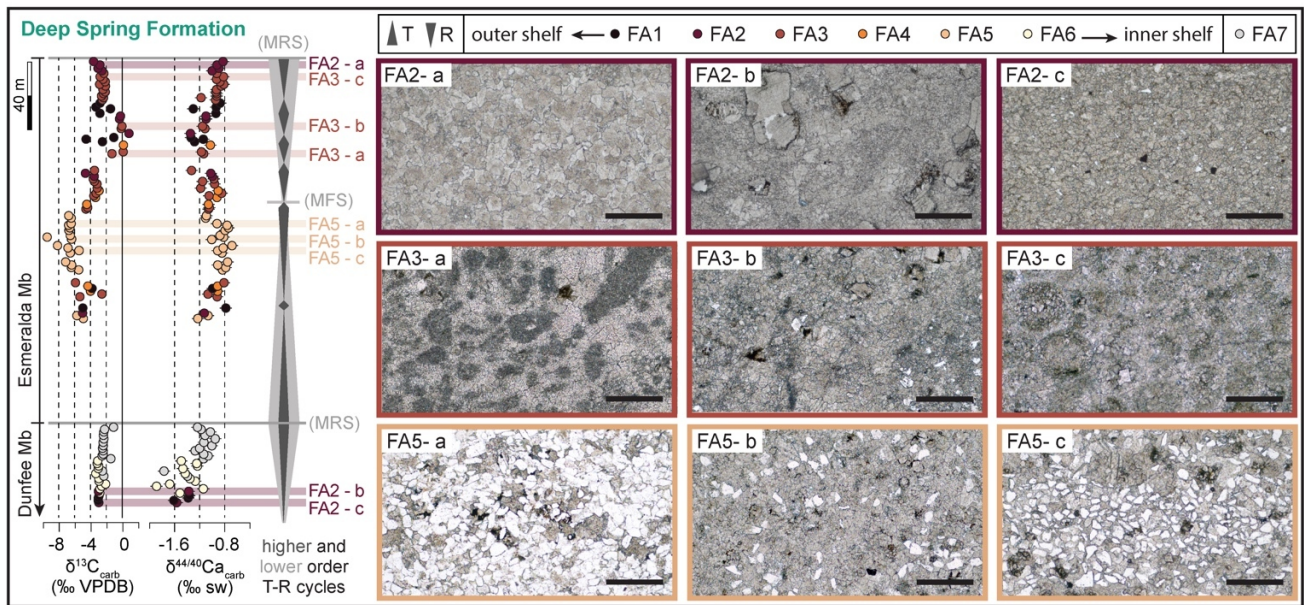


Figure 5. Stratigraphic variation of $\delta^{13}\text{C}$ and $\delta^{44/40}\text{Ca}$ from the Deep Spring Formation, colored by facies associations (FA) and integrated with sequence stratigraphy, with referenced thin-section photomicrographs of FA2, FA3, and FA5. Throughout the Esmeralda Member, $\delta^{13}\text{C}$ and $\delta^{44/40}\text{Ca}$ values vary within facies associations, although FA5 typically records low ($<-6\text{‰}$) $\delta^{13}\text{C}$ values and high ($>1.0\text{‰}$) $\delta^{44/40}\text{Ca}$ values. No relationship between carbonate fabric preservation and $\delta^{13}\text{C}$ or $\delta^{44/40}\text{Ca}$ values is observed across facies associations. For facies associations, refer to [table 1](#). T = transgressive, R = regressive. In photomicrographs, black bar = 500 μm . Top row: photomicrographs of FA2-a, FA2-b, and FA2-c (neomorphic spar), together displaying different levels of coarseness. Middle row: photomicrographs of FA3-a (peloidal cement), FA3-b (neomorphic spar with a small proportion of quartz grains), and FA3-c (ooid relics replaced by microspar), together displaying different degrees of fabric preservation. Bottom row: photomicrographs of FA5-a, FA5-b, and FA5-c (quartz grains in a microspar matrix), together displaying different proportions of quartz grains. The $\delta^{13}\text{C}$ data from Mount Dunfee, Cerro Rajón, and Echo Canyon are from Smith et al. (2016), Hodgins et al. (2021), and Smith et al. (2023), respectively. MRS=maximum regressive surface; MFS=maximum flooding surface.

that this interval was dominated by primary aragonite precipitation and that this mode of carbonate precipitation persisted through most of the Cambrian before gradually transitioning to a calcite sea (Wei et al., 2022).

It is unknown how much time is captured by the strata from which we report $\delta^{44/40}\text{Ca}$ variation. However, because $\delta^{44/40}\text{Ca}$ values vary by $\geq 0.4\text{‰}$ at all sites studied (figs. 1B and 2), the $\delta^{44/40}\text{Ca}$ variation cannot be entirely attributed to perturbations to global calcium cycle. It instead must be controlled, at least in part, by local changes in primary carbonate mineralogy and/or carbonate recrystallization and neomorphism.

5.1.2. Influence of local controls on calcium isotope fractionation

The $\delta^{44/40}\text{Ca}$ values presented here fall within the range of values predicted for either (1) a local change in conditions that control the dominant mineralogy of a platform during initial carbonate precipitation or (2) a transition between fluid-buffered and sediment-buffered conditions (with respect to calcium) during diagenesis (Ahm et al., 2018; Gussone et al., 2005). The relationship between Sr/Ca ratios and $\delta^{44/40}\text{Ca}$ values observed from limestones from the

Deep Spring Formation is consistent with either scenario ([fig. 2B](#)).

Mineralogy and precipitation rate are the primary controls on the Sr/Ca ratio and calcium isotope fractionation between CaCO_3 and seawater, where aragonite (versus calcite) precipitation and high (versus low) precipitation rates are associated with greater depletion in ^{44}Ca and higher Sr/Ca ratios (Fantle & DePaolo, 2007; Gussone et al., 2005; Tang et al., 2008). During neomorphism or recrystallization of carbonate minerals, precipitation rates do not appreciably fractionate calcium isotopes; however, during primary precipitation, they result in an $\sim 1.5\text{‰}$ average offset between aragonite and seawater $\delta^{44/40}\text{Ca}$ values and an $\sim 0.9\text{‰}$ average offset between calcite and seawater $\delta^{44/40}\text{Ca}$ values (Fantle & DePaolo, 2007; Gussone et al., 2005; Tang et al., 2008). One consequence of these fractionation effects is that fluid-buffered $\delta^{44/40}\text{Ca}$ values are readily distinguishable from sediment-buffered $\delta^{44/40}\text{Ca}$ values, with the more fluid-buffered values being closer to 0‰ and the more negative $\delta^{44/40}\text{Ca}$ values representing sediment-buffered values. As the extent of alteration under fluid-buffered conditions increases, carbonate $\delta^{44/40}\text{Ca}$ will approach fluid $\delta^{44/40}\text{Ca}$ values — a record is “fully” fluid-buffered when carbonate values match fluid values. Fluid- and sediment-buffered conditions occur along a con-

Table 1. Carbonate facies assignments for the top of the Dunfee Member and Esmeralda Member of the Deep Spring Formation at Mount Dunfee.

Carbonate facies associations		Description	Depositional Environment
FA1	Thin-bedded limestone	Thin-bedded (<30 cm) lime mudstone beds.	Outer shelf
FA2	Thin-bedded limestone with siltstone partings and interbeds	Thin-bedded (<30 cm) lime mudstone beds with thin siltstone interbeds and partings. Limestone beds occasionally display low-angle cross-lamination.	Outer shelf to shelf, oscillating around fair-weather wave base
FA3	Oolitic limestone	Thin- (<30 cm) to thick-bedded (1m) oolitic lime wackestone, packstone, and grainstone beds with occasional minor siltstone partings.	Carbonate shoal
FA4	Limestone stromatolites	Inclined columnar and broad hemispheroidal stromatolites preserved as limestone. Stromatolites appear patchily among recessive, lime wackestone, packstone, and grainstone beds.	Shelf
FA5	Sandstone with limestone lenses and interbeds	Thin-bedded (<30 cm) fine- to medium-grained quartz-rich sandstones with sandy lime wackestone, packstones, and grainstone interbeds and lenses.	Shelf to inner shelf with high sediment influx
FA6	Limestone mudchip debris flow	Thin-bedded (<30 cm) to massive lime mudstones with intraclast mudchip debris flows.	Inner shelf
FA7	Recrystallized dolostones	White to pink massive recrystallized dolostone beds. Occasionally, dolomitization appears to patchily affect the massive bed (uneven dolomitization front).	NA (post-depositional)

tinuum, with no defined cutoffs between the two end members. Similarly, strontium partitioning during diagenesis versus primary precipitation results in diagenetic minerals (dolomite, low-Mg calcite) that are Sr-poor relative to precursor minerals (aragonite, high-Mg calcite) (Tang et al., 2008).

5.2. Influence of diagenesis on $\delta^{44/40}\text{Ca}$, $\delta^{18}\text{O}$, and $\delta^{26}\text{Mg}$ records

The Dunfee Member of the Deep Spring Formation has the most negative $\delta^{44/40}\text{Ca}$ values of any of the studied samples, recording evidence of the most sediment-buffered diagenetic conditions (figs. 1B and 2). Our findings are complementary with previous studies that have shown how both fluid- and sediment-buffered conditions can lead to the preservation of primary $\delta^{13}\text{C}$ values depending on the depositional settings. Platform sediments that have been transported into deeper environments with little fluid flow (diffusion dominated), are argued to maintain their geochemical compositions in a sediment-buffered environment (Ahm et al., 2021; Husson et al., 2015). However, sediments deposited in shallow water environments can also preserve the $\delta^{13}\text{C}$ values of platform DIC if they undergo early fluid-buffered marine diagenesis by seawater that is not significantly evolved from its original composition (Crockford et al., 2021). In the Deep Spring, lower Wood Canyon, and La Ciénega formations the range in $\delta^{44/40}\text{Ca}$ values vary between the three sites (figs. 1B and 2). The $\delta^{44/40}\text{Ca}$ values within the BACE specifically also vary, despite consistency in the shape and magnitude of the $\delta^{13}\text{C}$ excursion. These variations in $\delta^{44/40}\text{Ca}$ are interpreted to record fluctuating diagenetic conditions, with the more positive $\delta^{44/40}\text{Ca}$ values recording more fluid-buffered di-

agenetic alteration. Of the three sections, the La Ciénega $\delta^{44/40}\text{Ca}$ values are the most elevated ($> -1.2\text{‰}$) indicative of persistent fluid-buffered alteration, consistent with the observed dolomitization of these strata (figs. 1B and 2). The range of $\delta^{44/40}\text{Ca}$ values from the lower Wood Canyon and Deep Spring formations are more variable, showing both fluid- and more sediment-buffered intervals (fig. 2).

5.2.1. Early marine dolomitization

The dolostones of the La Ciénega and lower Wood Canyon formations record geochemical signatures indicative of early marine dolomitization. The preservation of high ($> -0.8\text{‰}$) $\delta^{44/40}\text{Ca}$ values over some of the lower Wood Canyon Formation and throughout the La Ciénega Formation implies high degrees of alteration of $\delta^{44/40}\text{Ca}$ values in reaction with seawater, analogous to that documented to occur during early marine dolomitization in the Bahamas (Higgins et al., 2018). Additionally, in each formation, dolostones record relatively stable $\delta^{18}\text{O}$ values that are $\sim 30\text{‰}$ greater than Cambrian seawater $\delta^{18}\text{O}$ estimates ($\sim -36.3\text{‰}$ relative to VPDB; Hearing et al., 2018), which is consistent with oxygen isotope fractionation between seawater and dolomite at temperatures $< 40^\circ\text{C}$ (Horita, 2014; figs. 1B and 4A). Because $\delta^{18}\text{O}$ records are sensitive to diagenetic overprinting, the preservation of a relatively low-temperature signal implies dolomitization occurred prior to deep burial.

The low $\delta^{26}\text{Mg}$ values and high $\delta^{44/40}\text{Ca}$ values of the La Ciénega Formation further support geochemical stabilization during early marine dolomitization. In dolostones, $\delta^{26}\text{Mg}$ values can vary depending on magnesium availability during dolomitization. Generally, dolomites are depleted in ^{26}Mg relative to the dolomitizing fluid due to magnesium isotope fractionation during dolomite forma-

tion (fractionation factor of ~2‰, Fantle & Higgins, 2014; Higgins & Schrag, 2010). In a Mg-poor system, however, dolomite can be variably enriched in ^{26}Mg relative to a Mg-rich system due to Rayleigh-type distillation of the pore-fluid (Blättler et al., 2015). Over much of the La Ciénega Formation, $\delta^{26}\text{Mg}$ values are low and scattered between -2.3 and -1.7‰. Moreover, a coupling of $\delta^{44/40}\text{Ca}$ and $\delta^{26}\text{Mg}$ values is observed in strata that record the BACE. There, $\delta^{26}\text{Mg}$ values decrease by ~0.3‰ while $\delta^{44/40}\text{Ca}$ values increase by ~0.4‰ (figs. 1B and 4B), suggesting a shift in local environmental conditions that favor more fluid-buffered diagenesis with respect to both elements (e.g., greater advective flux of dolomitizing fluids).

Several mechanisms have been modeled to drive early, extensive dolomitization of carbonate platforms (e.g., Kaufman & Knoll, 1995; Whitaker et al., 2004). Some of these have distinct predictions for the evolution of dolomitizing fluid chemistries and, consequently, carbonate geochemical records (e.g., Riechermann et al., 2023). However, because initial platform geometries and paleoenvironmental conditions are not independently known for the La Ciénega and lower Wood Canyon formations, the likelihood of different dolomitization models cannot be meaningfully interrogated with the data presented. Nonetheless, we suggest that dolomitization models that involve the advection or convection of seawater or seawater-like fluids at temperatures <40°C (e.g., brine reflux, Kohout convection) are most consistent with the seawater-buffered records preserved in the La Ciénega and lower Wood Canyon Formations. In such models, the seawater flux will vary within the platform, influenced by platform geometry and lithologies (Al-Helal et al., 2012; Caspard et al., 2004; G. D. Jones & Xiao, 2005). We suggest that these controls on fluid flow may explain the differences in the extent of $\delta^{44/40}\text{Ca}$ alteration observed from the La Ciénega and lower Wood Canyon formations.

Evidence for partial dolomitization is also preserved in the top ~40 m of the Dunfee Member of the Deep Spring Formation, however, in these strata meteoric diagenesis likely also influenced the geochemistry. A transition toward fluid-buffered $\delta^{44/40}\text{Ca}$ values is recorded towards the top of the Dunfee Member, with dolostone $\delta^{44/40}\text{Ca}$ values stabilizing at ~-1.1‰, ~20 m below the top contact (fig. 1B). These values are lower than those predicted for fully seawater-buffered carbonates, given the -0.7‰ values reported from the La Ciénega Formation. This suggests meteoric fluids during dolomitization affected the upper Dunfee Member (Holmden et al., 2012; Riechermann et al., 2023). The presence of a karstic dissolution surface at the base of the Esmeralda Member is consistent with this interpretation. Additionally, the $\delta^{18}\text{O}$ values of the Dunfee Member are between -25‰ and -10‰, with the coarsely recrystallized dolostones stably recording $\delta^{18}\text{O}$ values of ~-15‰, depleted compared to dolostones of the lower Wood Canyon and La Ciénega formations (fig. 1B). While the mixing of meteoric and marine waters has been suggested to result in chemically favorable conditions for dolomite formation, kinetic barriers and the requirement of a significant Mg^{2+} source are argued to prevent significant dolomitization in

mixing zones (Hardie, 1987), consistent with the observation that patchy dolomitization is limited to the upper part of the Dunfee Member.

While there is petrographic evidence for later stage dolomitization of the upper Wood Canyon being associated with deep burial (Corsetti et al., 2006), our data suggest that major element carbonate geochemistry, like $\delta^{44/40}\text{Ca}$ and $\delta^{13}\text{C}$, was mostly resistant to these events. For example, dolostones from the top of the Dunfee Member of the Deep Spring Formation at Mount Dunfee preserve nonplanar petrographic textures (fig. S3), which are traditionally interpreted as evidence of crystal growth at >50 °C temperatures associated with burial (Sibley & Gregg, 1987; Woody et al., 1996), although recent work suggests they also may occur at low temperatures (Ryan et al., 2023). Similarly, petrographic data from the upper Wood Canyon Formation at Titanother Canyon indicates ooid silicification predated dolomitization (Corsetti et al., 2006), which was used to suggest that, at that locality, the Wood Canyon Formation was extensively buried prior to the introduction of dolomitizing fluids. It is, however, unclear whether the lower Wood Canyon Formation at Echo Canyon shares this dolomitization history. Regardless, because the dolostone $\delta^{44/40}\text{Ca}$ and $\delta^{26}\text{Mg}$ values presented here are consistent with the stabilization of geochemical records following early dolomitization, we suggest that dolomitization of the dolostone strata in the Wood Canyon and La Ciénega formations occurred early, likely driven by platform brine reflux, and major element dolostone geochemistry was mostly unaffected by later stage recrystallization.

5.2.2. Interpreting the $\delta^{18}\text{O}$ records

The $\delta^{44/40}\text{Ca}$ values, $\delta^{18}\text{O}$ values, and interpreted dolomitization histories of the La Ciénega and lower Wood Canyon formations are consistent with these sites recording seawater values during very early diagenesis. The $\delta^{18}\text{O}$ values in the Deep Spring Formation are significantly lower than those at the other two sites (figs. 1B and 4A). These low $\delta^{18}\text{O}$ values of the Deep Spring Formation could either indicate the resetting of the $\delta^{18}\text{O}$ record with meteoric fluids (plausible for the Dunfee Member which is beneath an exposure surface) and/or at high temperatures. The likely potential for burial diagenesis to reset $\delta^{18}\text{O}$ values but preserve $\delta^{44/40}\text{Ca}$ values in limestones has been suggested from modern and ancient environments, assuming sediment-buffered conditions with respect to calcium and carbon during carbonate burial (Ahm et al., 2018, 2021). Thus, for the Esmeralda Member of the Deep Spring Formation, we interpret a scenario in which limestones underwent partial diagenetic stabilization during early marine diagenesis. Final diagenetic stabilization occurred during burial, at which time closed-system conditions with respect to calcium and carbon prevailed. For the Dunfee Member of the Deep Spring Formation – which shows sedimentological evidence for exposure – we suggest that the lower $\delta^{18}\text{O}$ values could have resulted from meteoric fluids and/or burial diagenesis. This model is consistent with $\delta^{238}\text{U}$ isotope data from the upper Dunfee Member, which covary with $\delta^{44/40}\text{Ca}$ data (Chanchai et al., 2026).

5.3. Influence of early marine diagenesis on $\delta^{13}\text{C}$ records

While $\delta^{44/40}\text{Ca}$, $\delta^{18}\text{O}$, and $\delta^{26}\text{Mg}$ proxy records from the three sections of Ediacaran–Cambrian carbonates in south-west Laurentia are consistent with variable alteration by post-depositional diagenetic fluids, we argue that the large scale $\delta^{13}\text{C}$ trends in these records reflect changes in primary platform seawater isotopic composition. Two lines of evidence suggest that the BACE reflects changes to seawater chemistry rather than marine diagenesis. First, in the Deep Spring and La Ciénega formations, $\delta^{13}\text{C}$ values approach $\sim 0\text{‰}$ following recovery from the BACE, regardless of diagenetic regime. Although the full expression of the recovery is not preserved in the Wood Canyon or La Ciénega formations, all three sites preserve $\delta^{13}\text{C}$ values that return to $\sim 0\text{‰}$. This recovery of the $\delta^{13}\text{C}$ values in the La Ciénega Formation is recorded in a fluid-buffered diagenetic regime, in which dolostones record $\delta^{13}\text{C}$ values of $\sim 0\text{‰}$ and $\delta^{44/40}\text{Ca}$ values of up to $\sim -0.7\text{‰}$ (fig. 1B). In contrast to this, the $\delta^{13}\text{C}$ values following the recovery of the BACE in the Deep Spring Formation are relatively sediment-buffered, with limestones recording $\delta^{13}\text{C}$ values of $\sim 0\text{‰}$ and $\delta^{44/40}\text{Ca}$ values as low as $\sim -1.3\text{‰}$ (fig. 1B). Diagenetic modeling suggests that seawater-buffered diagenesis will only result in $\delta^{13}\text{C}$ records that approximate sediment-buffered $\delta^{13}\text{C}$ values if the concentration and $\delta^{13}\text{C}$ composition of DIC in seawater is consistent between diagenetic and depositional environments (Lau & Hardisty, 2022). Thus, the preservation of $\delta^{13}\text{C}$ values of $\sim 0\text{‰}$ in strata interpreted to record different degrees of $\delta^{44/40}\text{Ca}$ alteration indicates that the chemistry and concentration of DIC in seawater did not substantially change between precipitation and neomorphism/recrystallization, implying that diagenesis occurred very soon after carbonate deposition.

Second, the Esmeralda Member of the Deep Spring Formation does not preserve features indicative of authigenic carbonate precipitation, suggesting that authigenic carbonate precipitating in the pore-space likely did not play a significant role in driving bulk carbonate $\delta^{13}\text{C}$ trends (e.g., Schrag et al., 2013). In outcrop, there is no evidence of infilled void space or authigenic carbonate nodules throughout the member. In thin-section, only one generation of spar is observed from samples that record the BACE (fig. 5), suggesting the presence of neomorphic spar between sand grains, rather than authigenic cements (Bathurst, 1972). It is also unclear if platform redox conditions would have facilitated the anoxic degradation of organic matter required for the precipitation of authigenic carbonate with low $\delta^{13}\text{C}$ values, given that the BACE is reported from platform carbonates (e.g., sandy limestones of FA5; fig. 5). Bulk carbonate $\delta^{13}\text{C}$ values reported from the Esmeralda Member are therefore interpreted to largely record the $\delta^{13}\text{C}$ composition of carbonate minerals originally precipitated from seawater. Collectively, these lines of evidence suggest that the BACE was not driven by changes in diagenesis regimes and authigenic mineral precipitation, but instead by changes to the terminal Ediacaran platform seawater DIC pool.

Although the large magnitude $\delta^{13}\text{C}$ trends are not interpreted to result from diagenetic overprinting, fluctuations

in the diagenesis regimes recorded at each site are interpreted to have minor ($<2\text{‰}$) effects on $\delta^{13}\text{C}$ records. In a scenario where the $\delta^{13}\text{C}$ values of pore-fluids and seawater are similar, aragonite-to-calcite neomorphism is still expected to have different effects on bulk carbonate $\delta^{13}\text{C}$ values under different diagenetic regimes (Ahm et al., 2025). In a sediment-buffered regime (with respect to carbon), secondary calcite is expected to preserve the $\delta^{13}\text{C}$ value of the primary aragonite. However, in a fluid-buffered regime, secondary calcite will record the $\delta^{13}\text{C}$ value of pore-fluid plus the fractionation factor associated with calcite. As aragonite fractionates $\delta^{13}\text{C}$ values more strongly ($\sim 2.7\text{‰}$) than calcite ($\sim 1\text{‰}$), the transformation of aragonite to calcite under fluid-buffered conditions in seawater with the same $\delta^{13}\text{C}$ value of DIC should result in a $\sim 1.7\text{‰}$ decline in the $\delta^{13}\text{C}$ value of the sediment (Romanek et al., 1992). This scenario matches observations from our study sites. In the top ~ 60 m of the Deep Spring Formation, there are fluctuations between more sediment-buffered ($\sim -1.3\text{‰}$) and seawater-buffered ($\sim -0.8\text{‰}$) $\delta^{44/40}\text{Ca}$ values, coinciding with a decrease in $\delta^{13}\text{C}$ values from $\sim -0.2\text{‰}$ to $\sim -2.5\text{‰}$ (fig. 1B).

5.4 Influences of relative sea-level change, facies, and volcanism

It is unclear what mechanism promoted fluid-buffered diagenetic alteration across the margin during the BACE event, although porous, shallow water carbonate platforms are environments inherently prone to this style of diagenesis. Sedimentation rate has been suggested to control diagenetic regimes in marine environments (e.g., Higgins et al., 2018; L. L. Nelson et al., 2022), but the role of sedimentation rate in shaping the geochemical records of the La Ciénega, Wood Canyon, and Deep Spring formations is unknown, given the few radiometric age constraints that currently exist for these strata (Hodgin et al., 2021; L. L. Nelson et al., 2023). On other ancient carbonate platforms, greater proximity to shoreline is observed to correspond to higher degrees of alteration of carbonate geochemical records (Busch et al., 2022, 2023; Higgins et al., 2018; D. S. Jones et al., 2020). A similar control may explain the transition toward seawater-buffered $\delta^{44/40}\text{Ca}$ values corresponding with parts of the descending limb of the BACE that are associated with relative sea level falls. However, shallowing trends and facies associations do not consistently track the $\delta^{44/40}\text{Ca}$ variation recorded in the Esmeralda Member, which spans multiple transgressive-regressive cycles (fig. 5; Smith et al., 2023).

The physical properties of platform sediments can also determine diagenesis regime, as sediment porosity and permeability strongly affect platform circulation during early marine and meteoric diagenesis (e.g., Sanford et al., 1998). An increase in siliciclastic content among and within carbonate beds like that recorded alongside the BACE in the Deep Spring and La Ciénega formations may reflect such a control on diagenesis regime (figs. 1B and 5). However, the porosities and permeabilities of carbonate and siliciclastic sediments in marine diagenetic environments vary dramatically depending on factors like the size of carbonate grains and the extent of carbonate cementation at the time

of marine diagenesis (Ehrenberg & Nadeau, 2005; Enos & Sawatsky, 1981; Moore, 1989), which cannot directly be constrained from the sedimentary records of the Deep Spring and La Ciénega formations. Nonetheless, transitions between different facies associations over the Esmeralda Member do not correspond to any coherent trends in $\delta^{44/40}\text{Ca}$ values (fig. 5). Moreover, there is no strong evidence of a similar stratigraphic change in carbonate porosity or permeability in the lower Wood Canyon Formation, either because no change occurred or because later diagenesis (under sediment-buffered conditions) destroyed evidence of variation in carbonate facies.

We speculate that the increased fluid-buffered alteration across the BACE in southwestern Laurentia could be driven by the emplacement of volcanic dikes and sills that are genetically related to rift-related crustal thinning and faulting along the southwestern margin of Laurentia. A widespread increase in marine diagenesis could be explained by the circulation of hot seawater through carbonate platforms driven by dikes and sills in the subsurface. This mechanism is similar to Kohout driven convection but with a more powerful heat source. The geologic evidence for this hypothesis is limited, but not entirely speculative. Of the three sections studied, only the Caborca section has Ediacaran–Cambrian aged syndepositional basalts and volcanoclastics within it. The La Ciénega Formation also has the strongest evidence for early marine dolomitization (consistently fluid-buffered $\delta^{44/40}\text{Ca}$ values; fig. 2), implying that they were relatively proximal to the fluid source. Some of these basalts and volcanoclastics are found within the strata recording the BACE, but the thicker intervals of basalt and volcanoclastics occur in the Cerro Rajón Formation, the unit overlying the La Cienéga Formation (Barrón-Díaz et al., 2019; Hodgins et al., 2021; Stewart et al., 1984). Although there are no Ediacaran–Cambrian volcanics in the two other sections studied, there are basalt intervals within the latest Ediacaran–early Cambrian Stirling Quartzite and lower member of the Wood Canyon Formation farther north in Nevada (Smith et al., 2023; Stewart, 1970). This observation is consistent with the fact that the Deep Spring Formation is predominantly limestone while the Wood Canyon Formation is dolomitized, with slightly more sediment-buffered $\delta^{44/40}\text{Ca}$ values than in the La Ciénega Formation, implying a more distal relationship to the fluid source. This model for the emplacement for volcanic dikes and sills concomitant with the BACE is also consistent with positive Eu anomalies ($\text{Eu}/\text{Eu}^* > 1.5$) that occur at the nadir of the BACE in the Mount Dunfee and Cerro Rajón sections, potentially reflecting a regional input of hydrothermally sourced rare earth elements (Chanchai et al., 2026).

5.5. What does the BACE record?

Because the BACE is consistently preserved across >1000 kms and spanning multiple sequence boundaries in a continuum of more sediment- to more fluid-buffered diagenetic conditions in southwestern Laurentia, we suggest that this carbon isotope excursion primarily reflects secular trends in platform seawater DIC composition. Among the three studied successions, sequence stratigraphy and bios-

stratigraphy offer independent methods of correlation demonstrating that – even without precise radioisotopic constraints – the BACE occurs at generally the same time in all three sections. The carbonate strata recording the BACE in southwest Laurentia have clearly been altered through diagenesis. However, if these diagenetic fluids had played a major role in affecting the $\delta^{13}\text{C}$ of these strata, we would expect to see variable $\delta^{13}\text{C}$ values that correlate to the different diagenetic fingerprints. Instead, we document consistent $\delta^{13}\text{C}$ values preserved among the three study sites, and thus, the most parsimonious explanation of the $\delta^{13}\text{C}$ values of the BACE is that, in southwest Laurentia, they reflect platform seawater composition. It remains difficult to interpret whether this platform seawater reservoir was well mixed with the rest of the ocean.

If the BACE is recording a perturbation in a local DIC pool, then it could be explained by a shift in platform conditions. In modern evaporation pans, microbial mat productivity promotes disequilibrium between the concentrations of inorganic carbon in the seawater brine and the atmosphere, leading to the invasion of atmospheric CO_2 (Lazar & Erez, 1992). Kinetic isotope effects related to CO_2 hydration result in low ($\sim -9\text{‰}$) DIC $\delta^{13}\text{C}$ values (Lazar & Erez, 1992). Analogous processes have been hypothesized to occur on Neoproterozoic platforms where shelf-to-slope heterogeneity in the expression of large, negative $\delta^{13}\text{C}$ excursions is documented (Ahm et al., 2021; Busch et al., 2022). The BACE may similarly record an episode of enhanced platform productivity at the Ediacaran–Cambrian boundary. While such a scenario is highly speculative, high rates of productivity are associated with mass extinction and biotic turnover among primary producers in the Phanerozoic (Van De Schootbrugge & Gollner, 2013), implying a possible causal relationship between the BACE and potential ecological change in the terminal Ediacaran. One issue with interpreting the BACE in this way is the large spatial distance (~ 1000 km) over which the excursion is preserved in southwest Laurentia. Another issue is that there is no sedimentological evidence (e.g., evaporite pseudomorphs) that support the deposition of these strata within evaporitic environments. An arguably bigger issue is that there is little stratigraphic scatter in $\delta^{13}\text{C}$ values, a chemostratigraphic pattern that would be expected if the $\delta^{13}\text{C}$ values were the result of local isotope reservoir effects like CO_2 invasion.

In contrast to previous models proposed to explore triggers of the BACE and other large Neoproterozoic negative carbon isotope excursions, our data indicate a potential link between widespread changes in the $\delta^{13}\text{C}$ values of DIC and an increase in early fluid-buffered marine diagenesis of platform carbonates. As suggested above, one mechanism which could produce a widespread perturbation to the DIC pool and could be linked to an increase with marine fluid circulation in platform environments, involves magmatism associated with the rifting of the southwestern margin of Rodinia (Hodgins et al., 2021; Smith et al., 2023). In this model, rift-related dikes and sills intruded organic-rich strata and/or methane clathrates, resulting in the release of isotopically light carbon (Hodgins et al., 2021). While no physical evidence of organic-rich strata and/or

methane clathrates exists, there are late Ediacaran to early Cambrian basaltic sills in southwest Laurentia and a recently recognized Ediacaran–Cambrian large igneous province, the Wichita volcanics in southern Oklahoma; recent geochronology progress has demonstrated a broad temporal overlap between at least some of these igneous bodies and the Ediacaran–Cambrian boundary (Hodgin et al., 2021; L. L. Nelson et al., 2023; Smith et al., 2023; Wall et al., 2021). Additionally, in Caborca, the basaltic sills preserve chemistries consistent with crustal thinning during rifting (Barrón-Díaz et al., 2019; Tapia-Trinidad et al., 2022). Large igneous provinces have been repeatedly implicated in the generation of Phanerozoic negative $\delta^{13}\text{C}$ excursions and initiation of mass-extinction events (e.g., Berndt et al., 2023; Ruhl et al., 2011; Svensen et al., 2009; Wignall, 2001). The generation of the BACE and roughly coeval disappearance of Ediacaran faunal clades have been suggested to be analogous to these better constrained $\delta^{13}\text{C}$ excursions and mass extinctions (Hodgin et al., 2021; Smith et al., 2023).

6. CONCLUSIONS

Here, we assess the influence of marine diagenesis on the BACE along the western margin of Laurentia, the region from which it was first interpreted to mark the Ediacaran–Cambrian boundary. We find that the $\delta^{44/40}\text{Ca}$ values of the three studied sections record different degrees of mostly marine diagenetic alteration. The dolostone $\delta^{26}\text{Mg}$ and $\delta^{18}\text{O}$ values suggest that this diagenetic alteration occurred prior to significant burial. Despite the variability in $\delta^{44/40}\text{Ca}$ and interpreted type of diagenesis (i.e., more sediment- vs more fluid-buffered), the first order $\delta^{13}\text{C}$ chemostratigraphic trends are very similar between the three sections. There is smaller scale ($<2\text{‰}$) $\delta^{13}\text{C}$ variability among the sections, and, additionally, the Mount Dunfee section shows the most prevalent point-to-point variability among $\delta^{13}\text{C}$ data, while also showing the most stratigraphic variability in $\delta^{44/40}\text{Ca}$ data. Thus, we suggest that this smaller scale $\delta^{13}\text{C}$ variability can be attributed to stratigraphically variable patterns of early marine diagenesis which is in turn controlled by factors such as local base level and porosity. These changes in diagenesis were not responsible for the large, first-order, and regionally reproducible shifts in $\delta^{13}\text{C}$ across the BACE, which can instead be attributed to changes in platform seawater composition.

Ultimately, from the synthesis of $\delta^{13}\text{C}$, $\delta^{44/40}\text{Ca}$, $\delta^{26}\text{Mg}$, $\delta^{18}\text{O}$, and Sr/Ca records, we interpret a scenario in which seawater-buffered diagenesis occurred soon after the primary precipitation of aragonite, implying that the carbonates faithfully capture the $\delta^{13}\text{C}$ composition of platform

seawater DIC, which fluctuated to below-mantle values during the BACE. In this interpretation, seawater-buffered diagenesis did not generate a $\delta^{13}\text{C}$ excursion where one previously did not exist. Instead, it coincided with an interval of ^{13}C depletion in DIC in shallow marine environments and, potentially, in the global ocean.

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AUTHOR CONTRIBUTIONS

A.C.A., J.A.H., and E.F.S. designed the study. L.L.N. and E.F.S. collected the samples used in the study and provided geologic background context. M.C.L., J.W.T., and A.C.A. conducted the lab analyses for the study. M.C.L. wrote the initial draft of the manuscript with input from all coauthors. E.F.S. and J.A.H. raised funding for the work.

DATA AND SUPPLEMENTARY MATERIAL

The attached supplementary material provides extended methods for additional minor and trace element (Mn/Ca and U/Ca) data, as well as for calcium and magnesium isotope analyses. It provides additional figures presenting $\delta^{44/43}\text{Ca}$ and $\delta^{44/42}\text{Ca}$ and $\delta^{25/24}\text{Mg}$ and $\delta^{26/24}\text{Mg}$ values from the Deep Spring, Wood Canyon, and La Ciénega formations (fig. S1), Mn/Ca and U/Ca ratios from the Deep Spring, Wood Canyon, and La Ciénega formations (fig. S2), a photomicrograph displaying nonplanar dolomite texture from the top of the Dunfee Member of the Deep Spring Formation (fig. S3), and $\delta^{44/40}\text{Ca}$ and $\delta^{13}\text{C}$ values from the Deep Spring Formation color-scaled by facies associations and stratigraphic height (fig. S4).

Data are available through the Johns Hopkins Research Data Repository at <https://doi.org/10.7281/T1/IM9BZ1>.

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SUPPLEMENTARY MATERIALS

Lonsdale_laurentia_supplement_AJS

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