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Deconstructing the Lomagundi-Jatuli Carbon Isotope Excursion

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Abstract

The early to mid-Paleoproterozoic Lomagundi-Jatuli Excursion (LJE) is ostensibly the largest magnitude (approximately +5 to +30‰), longest duration (ca. 130–250 million years) positive carbon isotope excursion measured in carbonate rocks in Earth history. The LJE has been attributed to large nutrient fluxes, an increase in the size of the biosphere, a reorganization of the global carbon cycle, and oxygenation of the atmosphere. However, significant debate remains about its genesis, synchroneity, global-versus-local extent, and role in atmospheric oxygenation. Here we review existing models and mechanisms suggested for the LJE and analyze a compilation of ~9,400 $\delta^{13}C_{carb}$ and associated contextual data. These data call into question the interpretation of the LJE as a globally synchronous carbon isotope excursion and suggest that any model for the LJE must account for both the absence of a clearly defined initiation and termination of the excursion and a facies-dependent expression of ^{13}C-enrichment.

- The Lomagundi-Jatuli Excursion (LJE) continues to challenge current understandings of the carbon cycle.
- Understanding this excursion is critical for reconstructing biogeochemical cycles and atmospheric oxygenation through Earth history.

- Some evidence indicates local rather than global changes in $\delta^{13}C_{DIC}$ and raises the possibility of asynchronous, local excursions.
- Resolving whether the LJE was globally synchronous or asynchronous is essential for discriminating between different models.

1. INTRODUCTION

Over geologic time, the carbon isotope composition of marine carbonate minerals and rocks ($\delta^{13}C_{carb}$) has generally fluctuated near 0 permil (‰, parts per thousand relative to the Vienna Pee Dee Belemnite [VPDB] standard). Detailed study of sedimentary carbonate mineral and rock $\delta^{13}C_{carb}$ spanning Earth history has revealed many important perturbations from this baseline, both positive and negative, and varying in duration from short (hundreds of thousands of years or less) to very long (tens to hundreds of millions of years) (Knoll et al. 1986, Derry et al. 1992, Cox et al. 2016, Metzger et al. 2020). Demonstration that these carbon isotope excursions can be globally synchronous or near-synchronous (Swanson-Hysell et al. 2015, MacLennan et al. 2018, Metzger et al. 2020, Rooney et al. 2020; but see Melchin & Holmden 2006, Swart 2008, Smith & Swart 2022) has led to their widespread use as a tool for chemostratigraphic correlation and thus the construction of a framework for the relative timing of events throughout Earth history (Cramer & Jarvis 2020). This is particularly true for the Proterozoic [2.5 to 0.54 Ga (billion years ago)], where isotopic excursions have been identified in successions that can have absent, sparse, or contentious geochronological and biostratigraphic age constraints (McKenzie et al. 2013, Riedman et al. 2021). Further, because the $\delta^{13}C_{carb}$ of marine carbonate rocks is thought to track the balance of fluxes in the global carbon cycle, these data have become foundational in reconstructing the evolution of Earth's surface environments throughout geological time.

The Lomagundi-Jatuli Excursion (LJE) may be the largest, longest positive carbon isotope excursion measured in carbonate rocks over Earth history, typically recording $\delta^{13}C_{carb}$ values of approximately +5 to +10‰ (and up to +30‰) and potentially lasting ca. 100–250 million years (**Figure 1**). The LJE and Great Oxidation Event (GOE) (see the sidebar titled The Great Oxidation Event) have been broadly associated with postulated changes in the size and composition of the biosphere, nutrient fluxes, volcanic degassing, expansion and contraction of the seawater sulfate reservoir, the marine carbon cycle, and changes in tectonics and sedimentary recycling (Bekker & Holland 2012, Planavsky et al. 2012, Scott et al. 2014, Blättler et al. 2018, Crockford et al. 2019, Hodgskiss et al. 2019a, Eguchi et al. 2020, Hao et al. 2021). Nonetheless, there remains significant debate on even the most fundamental aspects of the LJE, such as whether it was a globally synchronous event (Bekker et al. 2006, Bekker & Holland 2012, Mayika et al. 2020), what the underlying causes were (Karhu & Holland 1996, Hayes & Waldbauer 2006, Bekker & Holland 2012, Eguchi et al. 2022), and whether it can even be considered a global carbon isotope excursion in the traditional sense (Mayika et al. 2020, 2021; Bekker et al. 2021; Prave et al. 2022). Simply put, if the elevated $\delta^{13}C_{carb}$ values characteristic of the LJE are representative of the oceanic reservoir of dissolved inorganic carbon (DIC), the sum of dissolved CO_2, HCO_3^-, and CO_3^{2-}, the excursion's magnitude is so large and duration so long that interpreting it within existing frameworks for the carbon cycle is extremely difficult (Aharon 2005, Hayes & Waldbauer 2006). A robust understanding of the nature of the carbon isotope excursion termed the LJE is therefore essential for exploring any potential significant changes occurring in the Earth system ca. 2.3–2.0 Ga and, more broadly, understanding the evolution of biogeochemical cycles through Earth history.

$\delta^{13}C$: the ratio of ^{13}C to ^{12}C in a sample relative to a standard (VPDB for carbonates): $\delta^{13}C_{carb} = \left(\frac{^{13}C/^{12}C_{sample}}{^{13}C/^{12}C_{standard}} - 1\right) * 1{,}000$

Here we compile 9,410 published $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ data, plus contextual information, for sedimentary carbonate rocks that span this pivotal time in Earth history during which both

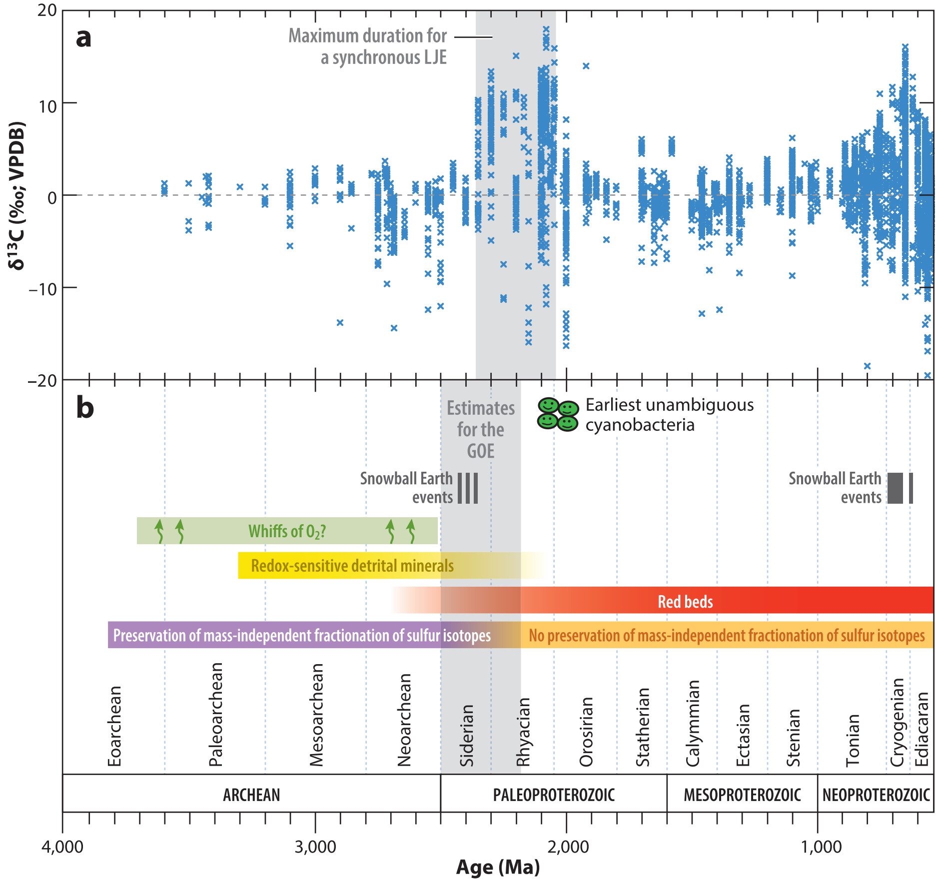


Figure 1

An overview of the $\delta^{13}C_{carb}$ record from the Archean to Neoproterozoic and other important events in Earth history over this time interval. The broad overlap in timing between the Great Oxidation Event (GOE) and Lomagundi-Jatuli Excursion (LJE) (*gray bars*) has led to different hypotheses linking these events, as discussed in the main text. (*a*) The $\delta^{13}C_{carb}$ record is dominated by values near 0‰, with two notable intervals of extreme values during the early to mid-Paleoproterozoic (i.e., the LJE) and Neoproterozoic. The gray bar indicates the approximate maximum duration of the LJE in a globally synchronous scenario (Martin et al. 2013). Carbon isotope data are from Shields & Veizer (2002). (*b*) Important changes in Earth's surface environment, including the GOE (*gray bar*) (age estimates from Gumsley et al. 2017, Warke et al. 2020, Hodgskiss & Sperling 2021, and Poulton et al. 2021), low-latitude glaciations (i.e., Snowball Earth events) (Caquineau et al. 2018, Hoffman et al. 2021), the earliest unambiguous fossil cyanobacteria (Hodgskiss et al. 2019b), and whiffs of oxygen during the Archean (Ostrander et al. 2021). The disappearance of redox-sensitive detrital minerals such as detrital pyrite and uraninite, the disappearance of mass-independent fractionation of sulfur isotopes, and the appearance of red beds and mass-dependent fractionation of sulfur isotopes are collectively interpreted to represent a shift toward a more oxidizing atmosphere (reviewed in Hodgskiss & Sperling 2021).

the GOE and LJE occurred. We review various models advanced over the past 50 years for explaining the LJE (and its potential relationship to the GOE) and use the compiled data and other accumulated observations to assess their viability. Our goal is to highlight the existing state of knowledge and potential future research directions with the hope of bringing the research community closer to an understanding of what remains one the biggest challenges presented by the sedimentary record.

THE GREAT OXIDATION EVENT

The Great Oxidation Event (GOE) was a pivotal time in Earth history that saw the rise of atmospheric oxygen levels above 10^{-5} present atmospheric levels. Evidence for the GOE has been drawn from a wide range of geological proxies [e.g., the appearance of red beds and disappearance of detrital pyrite and uraninite clasts in sedimentary units (reviewed in Johnson et al. 2014)] and geochemical proxies [e.g., the disappearance of mass-independent fractionation of sulfur isotopes in pyrite (Farquhar & Wing 2003)]. The exact timing of the GOE remains debated, however, with suggested intervals ranging from ca. 2.5 to 2.1 Ga (Gumsley et al. 2017, Warke et al. 2020, Hodgskiss & Sperling 2021, Poulton et al. 2021).

1.1. A Brief History of the Lomagundi-Jatuli Excursion

Paleoproterozoic sedimentary carbonate rocks significantly enriched in ^{13}C were first discovered in the Jatulian assemblages of Fennoscandia, with additional occurrences subsequently discovered elsewhere in Fennoscandia and Zimbabwe (Galimov et al. 1968, 1975; Schidlowski et al. 1975). Schidlowski et al. (1976) recognized the difficulties of interpreting these extremely positive $\delta^{13}C_{carb}$ values within existing geochemical and carbon cycle frameworks, a challenge that persists to this day. In the decades following those initial discoveries, Paleoproterozoic carbonate rocks with extreme ^{13}C enrichment have been found around the world (Karhu & Holland 1996, Maheshwari et al. 2010) (**Figure 2**).

The widespread occurrence of these sedimentary carbonate rocks with anomalous ^{13}C enrichments, broadly deposited between ca. 2.3 and 2.05 Ga, led many researchers to forward the idea that they collectively represent a global event (Karhu & Holland 1996, Melezhik et al. 1999) termed the Lomagundi-Jatuli Excursion after the Lomagundi (Zimbabwe) and Jatuli (Fennoscandia) localities (Karhu & Holland 1996, Melezhik & Fallick 1996, Melezhik et al. 1999). Of all the LJE-bearing regions globally, Fennoscandia and Gabon have risen to prominence as type localities for the LJE, in terms of sedimentology and geochemistry. In the former, the International Continental Scientific Drilling Program FAR-DEEP (Fennoscandia Arctic Russia-Drilling Early Earth Project) (see the sidebar titled FAR-DEEP) drilled new cores and combined these with other research and industrial drill cores to generate many scientific contributions toward understanding the LJE (Melezhik et al. 2013a–c). In Gabon, abundant industry drill cores through the Francevillian Group have made this another key succession for understanding the LJE (see the sidebar titled Francevillian Basin). Data from the Francevillian sections have resulted in publications on potential macrofossils, changes in ocean redox chemistry, and the carbon cycle (e.g., El Albani et al. 2010; Ossa Ossa et al. 2018, 2019; Préat & Weber 2019; Mayika et al. 2020). Important contributions for understanding the LJE have also come from the Transvaal Supergroup in South Africa and Paleoproterozoic successions in Brazil, Canada, the United States, and elsewhere (Bekker et al. 2006, Frauenstein et al. 2009, Maheshwari et al. 2010). A common thread between many studies of the LJE is a difficulty in explaining the origin of very positive $\delta^{13}C_{carb}$ signals that persisted for over 100 million years. We revisit these challenges below.

1.2. A Primer on Carbon Isotope Mass Balance and Implications for Oxygenation

The carbon isotope composition of carbonate rocks ($\delta^{13}C_{carb}$) is often interpreted through a simple isotope mass balance equation. In this framework, the carbon isotope composition of carbon input ($\delta^{13}C_{in}$) into Earth's surface environment (taken to be ~ -5‰ based on the assumed $\delta^{13}C$ of the mantle) from volcanism, metamorphic degassing, and chemical weathering is

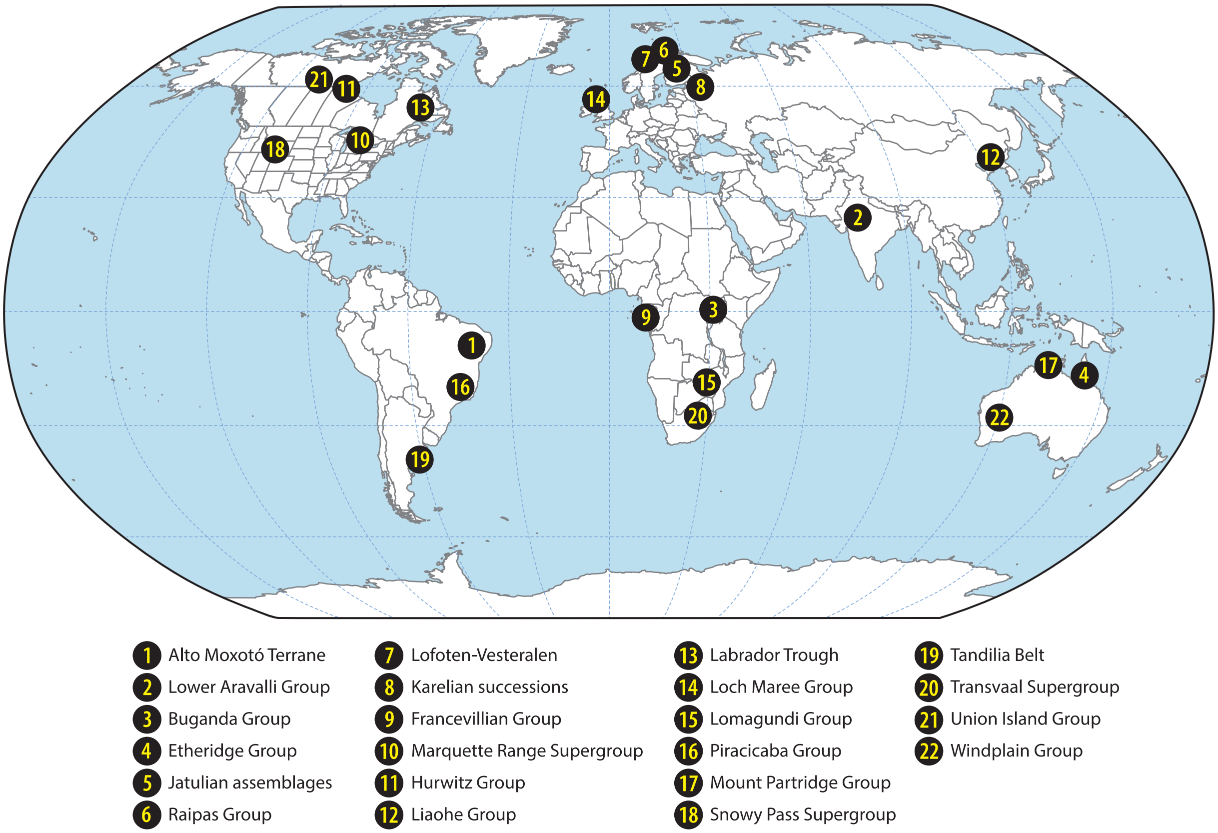


Figure 2

Locations of Lomagundi-Jatuli Excursion (LJE)-bearing and LJE-like successions (i.e., Paleoproterozoic rocks containing $\delta^{13}C_{carb}$ values greater than 5‰).

balanced by the fractions of carbon removed from Earth's surface environment as carbonate minerals (f_{carb}) and organic carbon (f_{org}) (see the sidebar titled f_{org}). Here it is generally assumed that $\delta^{13}C_{carb}$ directly reflects the inorganic carbon from which it is derived (typically assumed to be marine DIC). The carbon isotope fractionation between $\delta^{13}C_{carb}$ and $\delta^{13}C_{org}$ (termed ε) is assumed

FAR-DEEP

FAR-DEEP (Fennoscandia Arctic Russia-Drilling Early Earth Project) is a part of the International Continental Scientific Drilling Program that resulted in the collection of more than 3.6 km of drill core from Paleoproterozoic sedimentary basins in northeastern Russia. Collected material spans ca. 2.5 to 1.9 Ga and intersects Lomagundi-Jatuli Excursion (LJE) carbonate rocks, a Paleoproterozoic glacial unit, and an extremely organic-rich black shale overlying the LJE (Tulomozero, Umba, Kuetsjärvi, and Zaonega formations). These drill cores have been an invaluable archive for understanding the Paleoproterozoic Earth system and have resulted in 9 PhD theses, more than 40 published manuscripts, and 3 books (Melezhik et al. 2013a–c). FAR-DEEP drill cores are archived at the Geological Survey of Norway, where sampling programs continue to be organized for interested researchers.

FRANCEVILLIAN BASIN

The Francevillian Basin of Gabon is a well-preserved, approximately 2-km-thick package of sedimentary and volcanic rocks. These strata have been widely used as an archive of the ancient Earth system and have yielded important but controversial results, such as a potential two-step deoxygenation following the Great Oxidation Event/Lomagundi-Jatuli Excursion (LJE) (Ossa Ossa et al. 2018), macrofossils of multicellular organisms (El Albani et al. 2010, 2014, 2019), and a facies-dependent expression of the LJE (Mayika et al. 2020). Research on the Francevillian Basin has largely relied on drill core collected for industrial purposes, as surface outcrops are limited. Consequently, there is some debate on inferred depositional environments and the stratigraphic assignment of some strata, with implications for correlations and geochemical interpretations, causing disagreement among researchers (Ossa Ossa et al. 2019, Préat & Weber 2019, Bekker et al. 2021, Mayika et al. 2021). Further, the lack of robust age constraints hinders direct comparison of chemostratigraphic trends against other LJE strata.

to be ~25‰, which is the carbon isotope discrimination exhibited by the enzyme responsible for fixing inorganic carbon into organic carbon, ribulose-1,5-bisphosphate carboxylase-oxygenase (RuBisCO). Mathematically, this carbon isotope mass balance is given by

$$\delta^{13}C_{in} = \delta^{13}C_{carb} * f_{carb} + \delta^{13}C_{org} * f_{org}, \quad 1.$$

where

$$f_{carb} + f_{org} = 1 \quad 2.$$

and, due to the isotopic fractionation during organic carbon formation,

$$\delta^{13}C_{org} = \delta^{13}C_{carb} - \varepsilon. \quad 3.$$

There are many important assumptions implicit in this simplified carbon isotope mass balance. First, there is an assumption that carbon is removed from Earth's surface only through the burial of carbonate rocks or organic carbon. Second, this assumes that $\delta^{13}C_{carb}$ faithfully records the isotopic composition of ambient DIC. Third, it assumes that we can average processes that we know partition carbon isotopes among the various surface reservoirs, for example, changes in the air-sea gas exchange between CO_2 in the atmosphere and ocean, or precipitation of carbon into different carbonate polymorphs and minerals (e.g., calcite, aragonite, dolomite). Finally, this carbon isotope mass balance also assumes that there have not been significant changes in carbon isotope fractionation associated with different types of RuBisCO (i.e., ε is constant) and that $\delta^{13}C_{in}$ has not changed appreciably through Earth history. With these assumptions in mind, combining

f_{org}

f_{org} is the ratio of organic carbon to total carbon buried from Earth's surface environment. This is canonically interpreted as the primary lever on seawater $\delta^{13}C_{DIC}$ and therefore $\delta^{13}C_{carb}$. To maintain the ~0‰ values that are prevalent through much of Earth history, f_{org} is thought to have generally ranged from ~0.2 to 0.3 (or 20–30% of total carbon burial). In this framework, the very positive $\delta^{13}C_{carb}$ values characteristic of the Lomagundi-Jatuli Excursion may represent extremely elevated f_{org} (i.e., a significant increase in the burial or organic carbon relative to carbonate carbon). However, some researchers believe that such a large, sustained shift in f_{org} is unlikely and argue that this framework is inadequate for explaining such a large magnitude, long duration carbon isotope excursion.

measurements of $\delta^{13}C_{carb}$ with values for $\delta^{13}C_{in}$ and ε, f_{org} can be calculated:

$$f_{org} = \frac{(\delta^{13}C_{carb} - \delta^{13}C_{in})}{\varepsilon}. \quad 4.$$

It follows from Equation 4 that when $\delta^{13}C_{in}$ and ε are assumed to be constant, then $\delta^{13}C_{carb}$ is directly proportional to f_{org}. Quantifying and documenting shifts in f_{org} is important because burying organic carbon removes reduced carbon (and preferentially ^{12}C) from Earth's surface environment, causing oxygenation. It is likely through this organic carbon burial that significant amounts of oxygen (O_2) gas came to exist in Earth's atmosphere:

$$CO_2 + H_2O \underset{\text{light}}{\rightarrow} CH_2O + O_2. \quad 5.$$

The framework of this simple isotope mass balance is how $\delta^{13}C_{carb}$ data sets over Earth history have largely been interpreted.

The extremely positive $\delta^{13}C_{carb}$ values of the LJE were initially interpreted within this framework as representing a significant increase in f_{org}, at least on a local scale (Schidlowski et al. 1975, 1976). As the number of Paleoproterozoic localities with very positive $\delta^{13}C_{carb}$ grew, interpretations of the LJE shifted toward global rather than local increases in f_{org}, an overall increase in the organic carbon burial flux, and an associated, sustained increase of atmospheric oxygen levels (Karhu & Holland 1996, Melezhik et al. 1999). To explain global $\delta^{13}C_{carb}$ of +5 to +10‰ during the LJE requires f_{org} to increase from a historical average of 0.2 up to 0.4 to 0.6 and be sustained for hundreds of millions of years. Such an interpretation broadly aligns the LJE with evidence for atmospheric oxygenation and the GOE around this time interval, including the appearance of significant sulfate evaporite minerals and red beds, the disappearance of detrital pyrite and uraninite, and the cessation of mass-independent fractionation of sulfur isotopes (MIF-S) in sedimentary pyrite (Farquhar & Wing 2003, Johnson et al. 2014, Hodgskiss & Sperling 2021). However, robust constraints on the LJE's timing, duration, synchroneity, and cause(s) remain lacking. These uncertainties have instigated a significant divergence on the interpretation of the LJE and more broadly on the interpretation of the carbon isotope composition of carbonate rocks ($\delta^{13}C_{carb}$) over geological time.

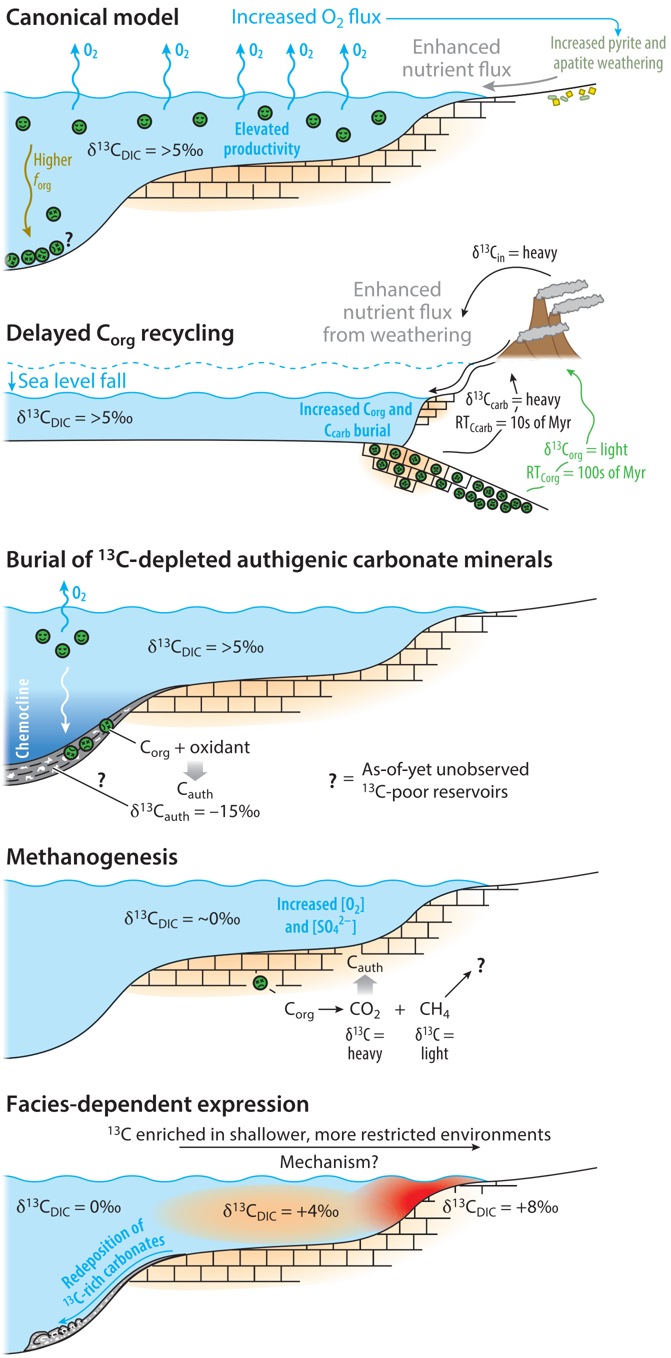
2. SUGGESTED MODELS FOR THE LOMAGUNDI-JATULI EXCURSION

A wide range of models and mechanisms have been proposed for the sustained very positive $\delta^{13}C_{carb}$ observed during the LJE. Importantly, these models differ in fundamental aspects, such as what phases sequester the excess ^{12}C and where these phases were deposited. Models also differ in terms of invoking global or local shifts in the $\delta^{13}C$ of DIC and thus whether the LJE represents a global perturbation to the carbon cycle. Finally, the proposed models differ on whether the LJE was a cause or an effect of atmospheric/oceanic oxygenation.

2.1. The Canonical Model

The canonical model for the LJE interprets the increase in $\delta^{13}C_{carb}$ as globally synchronous and representing a marked increase in f_{org} (**Figure 3**). Despite the broad adoption of this model, there remains significant uncertainty regarding what might have triggered and sustained such a large increase in f_{org}. Bekker & Holland (2012) suggested that an initial increase in atmospheric oxygen levels may have promoted the weathering of pyrite (FeS_2) and other sulfide minerals, producing sulfuric acid (H_2SO_4) that dissolved apatite [$Ca_5(PO_4)_3(F,Cl,OH)$] and other phosphate minerals, thereby resulting in an increased phosphorus flux to the oceans; under the assumption of a phosphorus-limited biosphere, an increased phosphorus flux would promote enhanced primary productivity and presumably organic carbon burial, increasing oxygen production and driving a

MODEL



EXPLANATION

A significant increase in primary productivity results in elevated f_{org} and therefore increased seawater $\delta^{13}C_{DIC}$. This may be driven by a positive feedback loop in which O_2 from primary producers weathers pyrite to produce sulfuric acid, which then weathers apatite to release phosphorus. This enhanced nutrient flux then increases primary productivity.

Enhanced volcanism during supercontinent breakup increases both C_{org} and C_{carb} burial (f_{org} is constant). Relative sea level fall focused C_{org} and C_{carb} burial in subduction zones. Subducted C_{carb} (^{13}C rich) had a short residence time in the mantle (tens of Myr), whereas C_{org} (^{13}C poor) resided deeper in the mantle for hundreds of Myr, resulting in a shift in volcanic C input (and ocean DIC) toward positive $\delta^{13}C$ values.

Remineralization of organic matter below a chemocline resulted in formation of ^{13}C-depleted authigenic carbonate minerals. Burial of these authigenic carbonates drove residual seawater $\delta^{13}C_{DIC}$ toward positive values.

An increase in availability of electron acceptors (e.g., O_2, SO_4^{2-}) associated with ocean oxygenation pushed methanogenesis deeper in the sediment column. Authigenic carbonates forming from porewaters were ^{13}C rich due to CO_2 from methanogenesis, rather than composition of the ocean DIC reservoir.

Water column $\delta^{13}C_{DIC}$ was strongly elevated in near-shore, inner shelf, intertidal, and sabkha environments but remained ~0‰ in open and deep marine environments. The occurrence of high $\delta^{13}C$ carbonates in some deep marine environments may be due to redeposition of sediments from shallower environments.

(Caption appears on following page)

Figure 3 (*Figure appears on preceding page*)

Different models proposed for the Lomagundi-Jatuli Excursion invoke a range of triggers, including elevated primary productivity due to a positive feedback loop, changes in carbon recycling, formation of authigenic carbonate, and the restriction of elevated $\delta^{13}C_{carb}$ to near-shore and restricted environments. Abbreviations: C_{auth}, authigenic carbon; C_{carb}, carbonate carbon; C_{DIC}, dissolved inorganic carbon; C_{in}, carbon influx; C_{org}, organic carbon; DIC, dissolved inorganic carbon; f_{org}, fraction of carbon buried as organic carbon; RT_{Ccarb}, residence time of carbonate carbon; RT_{Corg}, residence time of organic carbon.

positive feedback loop. Alternatively, increased primary productivity leading to increased f_{org} may have been driven by changes in nutrient fluxes including phosphorus but also iron and manganese, due to tectonic processes. Indeed, the LJE broadly coincides with important tectonic changes, such as a global tectono-magmatic lull ca. 2.36 to 2.23 Ga during which magma/mantle plume production and orogenic activity may have decreased (Condie et al. 2009, 2022). Campbell & Allen (2008) suggested that the formation and erosion of large orogenic belts throughout Earth history may have supplied significant amounts of nutrients, such as iron and phosphorus, and increased primary productivity and burial of organic matter. Alternatively, rather than nutrient delivery stimulating increased primary productivity, it has been suggested that environments suitable for organic carbon burial, specifically continental shelves, may have developed around this time. A significant increase in the subaerial extent of the continents and continental shelf area ca. 2.5 to 2.1 Ga has been interpreted based on triple oxygen isotope ratio data from shale and granite samples, potentially reflecting the widespread emergence of continents similar in aerial extent and hypsometry to the modern Earth (Bindeman et al. 2018, Bindeman 2021). Similarly, it has been proposed that large igneous provinces record a shift to increased subaerial exposure of continents from ca. 2.4 to 2.2 Ga (Liebmann et al. 2022).

The confluence of tectonic events likely associated with this time interval may have driven a dramatic combination of events that led to increased f_{org}, including an increase in nutrient fluxes to the oceans stimulating primary productivity and the creation/enlargement of environments conducive for organic carbon deposition (Bindeman et al. 2018, Bindeman 2021). This is particularly true given that environments with high sedimentation rates such as continental shelves have been shown to enhance organic matter burial (Hedges & Keil 1995, Hemingway et al. 2019). However, recent evidence suggests that the growth of continental crust and significant orogenesis occurred hundreds of millions of years earlier than the LJE (Tang et al. 2021), and indeed, there is much debate over the timing and extent of continental crust growth over the course of Earth history (Hawkesworth et al. 2019). In summary, these models for the LJE broadly suggest that tectonic mechanisms drove increases in f_{org} through a combination of increased nutrient fluxes and expansion of environments favorable for organic carbon burial, resulting in the production and accumulation of significant amounts of oxygen. However, the specific timing of these events remains a major source of uncertainty.

The burial of such a large amount of organic carbon driving increased $\delta^{13}C_{carb}$ predicts that the LJE corresponds to a major interval of oxygen production, and indeed, previous calculations have estimated the production of enormous amounts of oxygen over the LJE (Karhu & Holland 1996, Bekker & Holland 2012). This connection between the LJE and GOE has been inferred since some of the earliest definitions for the GOE, where it was suggested that the GOE was a ca. 400 million-year-long event, with a termination that is marked by $\delta^{13}C$ values of carbonate rocks returning to $\sim$0‰ (Holland 2002). Existing geochronological data indicate that oxygenation of the atmosphere, as constrained by the loss of MIF-S in the sedimentary record, occurred between ca. 2.5 and 2.2 Ga (Luo et al. 2016, Gumsley et al. 2017, Warke et al. 2020, Poulton et al. 2021), with confidence interval analyses suggesting oxygenation could have occurred as late as ca. 2.1 Ga (Hodgskiss & Sperling 2021). The canonical model requires that the LJE was a globally synchronous event, and although there are some regions that record both the disappearance of MIF-S (i.e., onset of the GOE) and a rise in $\delta^{13}C_{carb}$ (i.e., the onset of the LJE), straightforward interpretations of these records are made more difficult by generally sparse age constraints, different mineral records for each proxy, uncertainties in proxy interpretation, and an incomplete geological record (Martin et al. 2013, Poulton et al. 2021). Whether the LJE preceded, coincided with, or postdated the GOE is thus unclear.

The canonical model also struggles with an issue common to many models invoked to explain the LJE: There is generally little geological evidence for increased f_{org}. Specifically, there does not appear to be significant accumulation of organic-rich sedimentary rocks during the LJE. While some remarkably organic-rich strata have been observed in association with LJE carbonate rocks (e.g., Zaonega Formation, Russia; FB, FC, and FD formations, Gabon), these appear to have been deposited toward the end of the LJE or perhaps afterward. Another issue with the canonical model for the LJE is that there does not appear to be a trend toward more positive $\delta^{13}C_{org}$ that would be predicted if there was a large increase in f_{org} driving the large increase in $\delta^{13}C_{carb}$ (Karhu & Holland 1996; but see Bekker et al. 2008). These criticisms together with geochronological uncertainty have motivated the development of other models for the LJE.

2.2. Changes in Carbon Recycling to Break the Link to Oxygenation

Some researchers have investigated possible scenarios for the LJE that allow for a globally synchronous increase in $\delta^{13}C_{DIC}$ (and therefore $\delta^{13}C_{carb}$) with stable or only moderate increases in f_{org}, thereby avoiding the release of enormous amounts of oxygen and the issue that uniquely organic-rich sedimentary rocks are not widespread at this time. For example, Eguchi et al. (2020, 2022) used a model to suggest that elevated weathering of basaltic landmasses in the Paleoproterozoic would increase nutrient and alkalinity supply to the oceans, increasing deposition of both carbonate sediments and organic carbon (i.e., f_{org} remains approximately constant). As part of this model, mantle plumes during supercontinent breakup around this time may have caused relative sea level fall, focusing burial of carbonate minerals and organic carbon along subducting margins; carbonate carbon tends to volatilize at subarc depths and therefore has a mantle residence time of tens of millions of years, whereas organic carbon is volatized at greater depths and has a mantle residence time of hundreds of millions of years (Bouilhol et al. 2022). Therefore, subducted sediments would have preferentially released carbonate carbon back to Earth's surface system through devolatilization, shifting $\delta^{13}C_{in}$ and therefore $\delta^{13}C_{carb}$ to more positive values as a result (**Figure 3**). The occurrence of these processes over tectonic timescales may also explain a potential lag of ~200 million years between the onsets of the GOE (initial increase in f_{org}) and LJE (carbonate recycling driving higher $\delta^{13}C_{carb}$). However, mantle plume–driven supercontinent breakup has occurred other times throughout Earth history and does not otherwise result in carbon isotope excursions of the magnitude and duration of the LJE.

Other models have also been suggested to explain positive $\delta^{13}C_{carb}$ during the LJE without invoking large increases in f_{org}. Shields & Mills (2017) considered the flux of weathering of carbonate rocks and proposed that positive $\delta^{13}C_{carb}$ can, in some cases, be decoupled from oxygen production. The crux of this model is that carbonate minerals weather more readily than organic carbon in regimes characterized by high erosion rates (e.g., during tectonic uplift or orogenic events) and are subsequently reprecipitated in the oceans, thereby decreasing f_{org} and $\delta^{13}C_{DIC}$. Such scenarios allow for changing f_{org} without changing the absolute amount of organic carbon buried, thereby decoupling $\delta^{13}C_{carb}$ from atmospheric oxygen levels. Similarly, Miyazaki et al. (2018) constructed a carbon cycle model that allows for variations in the rate of sedimentary carbon weathering/recycling and coupled organic carbon oxidation to atmospheric oxygen levels. In this model, they proposed that weathering of recycled carbonate sediments with positive isotopic signatures would allow for very positive sedimentary carbonate $\delta^{13}C_{carb}$ with only moderate increases in f_{org}.

While these models may help to explain certain aspects of the LJE, they raise other questions about the mechanism for increased $\delta^{13}C_{carb}$. Ultimately the LJE is a unique carbon isotope excursion, whereas these tectonically driven increases in alkalinity delivery or carbonate weathering are relatively common throughout Earth history. It therefore remains unclear why these processes would have occurred uniquely during this time interval compared to the rest of Earth history.

2.3. Driving Positive $\delta^{13}C$ Through Sediment Column Processes

Other proposed mechanisms for the LJE invoke the formation of carbonate minerals within the sediment column via processes with strong carbon isotope fractionations. These mechanisms would allow for the preservation of high $\delta^{13}C_{carb}$ without impacting the overall fluxes in the global carbon cycle and therefore have minimal impact on f_{org} or atmospheric oxygen levels. These sedimentary processes include the formation of ^{13}C-rich carbonate sediments through methanogenesis, or ^{13}C-poor carbonate sediments through authigenic carbonate formation, and in many ways are two sides of the same coin; methane production and consumption strongly partition carbon isotopes so carbonate minerals that form during methanogenesis or methanotrophy would be isotopically unique. Indeed, the large difference between $\delta^{13}C_{methane}$ and other carbon reservoirs has led to methane production or oxidation being the suggested lever for nearly all $\delta^{13}C_{carb}$ excursions, both positive and negative, over geologic time.

2.3.1. Methanogenesis in the sediment column.

Chief among sediment column processes invoked for the formation of extremely ^{13}C-enriched carbonate minerals throughout much of Earth history is methanogenesis (**Figure 3**). As a product of microbial methanogenesis, methane is typically very ^{13}C depleted ($\delta^{13}C_{methane} = -50$ to $-100‰$), and regions or time intervals associated with enhanced methane production or storage would therefore have a residual DIC reservoir that is ^{13}C enriched, ultimately leading to higher $\delta^{13}C_{carb}$ formed from this reservoir. For example, Hayes & Waldbauer (2006) suggested that a deepening of the methanic zone in marine sediments during initial surface ocean oxygenation associated with the GOE would drive porewater DIC toward more positive $\delta^{13}C$ values. Early diagenetic carbonate minerals that source carbon from this porewater reservoir would therefore record a significant ^{13}C enrichment. We note that this mechanism can work only if carbonate mineral precipitation occurred with methane production; however, methane production typically lowers pH and, as a consequence, inhibits carbonate mineral precipitation (Soetaert et al. 2007). Conversely, anaerobic methane oxidation locally raises pH and promotes carbonate mineral precipitation (with very ^{12}C-rich carbonate minerals). That the ^{13}C-enriched carbonate derived from methane production would be globally preserved, rather than the thermodynamically favored ^{13}C-depleted carbonate minerals associated with methane oxidation, appears to be a conundrum. Indeed, there are no globally significant units with ^{13}C-depleted carbonate rocks found across the LJE that would be expected from the oxidation of the product methane. Further, sulfur isotope data from LJE carbonate rocks and sulfate evaporite minerals have been interpreted to indicate that methanogenesis was not an important control on $\delta^{13}C_{carb}$ during the LJE (Planavsky et al. 2012).

There are modern environmental analogs where in situ methane production leads to carbonate minerals forming with extremely positive $\delta^{13}C_{carb}$. For example, stromatolites growing in a modern methanogenic and brackish to hypersaline lagoon in Brazil record $\delta^{13}C_{carb}$ up to $+16‰$; these stromatolites have been interpreted to represent carbonate mineral precipitation coincident with locally significant methanogenesis (Birgel et al. 2015). On Pamanzi Island (Indian Ocean), a

AUTHIGENIC CARBONATE

Authigenic carbonate minerals precipitate in situ within the sediment column (in porewaters) or near the sediment-water interface. Carbon in these minerals is typically sourced from oxidation of organic matter or methane, generally resulting in strongly negative $\delta^{13}C_{carb}$. The $\delta^{13}C$ of modern authigenic carbonate minerals ranges from −60‰ to more than +20‰ (with very positive values typically attributed to methanogenesis), although most values are strongly negative. Authigenic carbonate precipitation has been estimated as only a small part of the modern carbonate burial flux (Bradbury & Turchyn 2019) but can be locally very important, such as in the Amazon River delta where it comprises 30% of total carbon burial (Aller et al. 1996).

redox-stratified saline lake has been proposed as an analog for restricted waterbodies in the Proterozoic (Cadeau et al. 2020). Due to intense primary productivity and subsequent organic carbon remineralization during methanogenesis in the sediment column, the $\delta^{13}C_{DIC}$ in the lake ranges from +11 to +13‰, and the mean $\delta^{13}C_{carb}$ of the sediment is +16‰. However, both of these analog environments are geographically small (lake surface areas of 16 and 1.8 km^2, respectively), and it is unclear whether such environments can be extrapolated to a global scale and persist over suggested depositional timescales for the LJE.

2.3.2. Burial of ^{13}C-depleted authigenic carbonate minerals. An alternative model for the LJE invokes widespread precipitation of authigenic carbonate minerals (see the sidebar titled Authigenic Carbonate) with very negative $\delta^{13}C_{carb}$, formed through oxidation of organic matter or methane within the sediment column. If authigenic carbonate mineral formation was widespread during the LJE, it would result in the sequestration of large amounts of ^{12}C in sedimentary carbonate minerals and therefore an increase in residual marine $\delta^{13}C_{DIC}$ (Higgins et al. 2009, Schrag et al. 2013). Variations in the formation and burial rates of these strongly ^{12}C-enriched authigenic carbonate minerals have been invoked to explain important aspects of the $\delta^{13}C$ record, such as the persistence of baseline marine $\delta^{13}C$ values near 0‰ through most of Earth history (Laakso & Schrag 2022), despite oxygen production rates that must have varied significantly. In this framework, the LJE would be explained, at least in part, by increased burial of authigenic carbonate minerals.

Although this model may be able to explain the LJE without invoking order-of-magnitude changes in burial fluxes of organic carbon (Laakso & Schrag 2022), it has not achieved widespread acceptance, primarily because direct evidence of enhanced authigenic carbonate mineral formation in the geological record is sparse, let alone during the LJE. The initial hypothesis of Higgins et al. (2009) implied that authigenic carbonate formation would be minimal in oxygenated oceans; this has been corroborated through modern sedimentary pore fluid database analyses (Bradbury & Turchyn 2019), although authigenic carbonate mineral formation can be locally significant, such as on the Amazon shelf, where it is estimated to constitute ∼30% of total carbon burial locally (Aller et al. 1996). Canfield et al. (2020) analyzed the $\delta^{13}C_{auth}$ of 541 siliciclastic rocks spanning ca. 840 to 540 Ma, found little ^{12}C-enriched carbonate, and concluded that although it is most correct to consider authigenic carbonate fluxes, they likely exert very little control over the carbon cycle from an isotopic mass balance perspective. For their part, Laakso & Schrag (2022) suggested that more intensive sampling of ancient deep-water sedimentary rocks may be necessary to fully ascertain the overall importance of the authigenic carbonate burial sink.

2.4. A Facies-Dependent Explanation for the Lomagundi-Jatuli Excursion

In contrast to models that have sought to reconcile very positive $\delta^{13}C_{carb}$ values with the global carbon cycle, there are models that question the underlying assumption that these $\delta^{13}C_{carb}$ values are representative of global seawater DIC rather than local/basin waters. Inherent in such models is the idea that ^{13}C-enriched carbonate rocks were not necessarily deposited synchronously. If the LJE carbonate rocks are not globally synchronous, then the deposition of ^{13}C-rich carbonate rocks at a given location may be influenced by local factors such as water mass restriction, changes in relative sea level, and air-sea gas exchange, in addition to whatever global changes were occurring within the broader Earth system at this time (**Figure 3**). The possibility that the LJE may be facies dependent or related to local conditions has long been suggested (e.g., Schidlowski et al. 1976). Melezhik et al. (1999) noted that some restricted, evaporitic environments in the LJE-bearing Tulomozero Formation (Russia) record strongly elevated $\delta^{13}C_{carb}$ values up to +17‰, compared to an inferred global ocean baseline of approximately +5‰. These elevated $\delta^{13}C_{carb}$ values were then interpreted to be the result of locally restricted and evaporitic conditions, high biological productivity, and possible methane production, mirroring conditions observed in some niche environments today (as described in Section 2.3.1).

Prave et al. (2022) compiled $\delta^{13}C_{carb}$ and depositional environment information from 14,943 carbonate sedimentary rocks (2,038 associated with the LJE) spanning ca. 3.0 to 1.0 Ga to better contextualize the anomalously ^{13}C-enriched carbonates associated with the LJE. Binning the samples into depositional environments of intertidal-coastal sabkha, nearshore-inner shelf, and open and deeper marine, their results indicate that open and deeper marine settings saw only a modest increase in $\delta^{13}C_{carb}$ (mean of 1.5 ± 2.4‰), within the range considered to be normal seawater. The nearshore-inner shelf environment saw a larger increase (mean of 6.2 ± 2.0‰), and intertidal-coastal sabkha saw the largest increase as well as the most variability (mean of 8.1 ± 3.8‰). Prave et al. (2022) also presented high-resolution $\delta^{13}C_{carb}$ chemostratigraphic data from the LJE-bearing Tulomozero and Zaonega formations, and they suggested that a sharp decrease of approximately 5‰ within the LJE coincides with an abrupt transition from nearshore-intertidal-coastal sabkha facies to open and deeper marine facies within the study locality, similar to a facies-$\delta^{13}C_{carb}$ relationship observed in the Francevillian Basin (Mayika et al. 2020). Although they do not suggest a specific mechanism for why very positive $\delta^{13}C_{carb}$ values would be restricted to near-shore environments, they note that facies-dependent variations of several permil have been observed in both ancient and modern carbonate sedimentary systems (e.g., Macdonald et al. 2009, Swart et al. 2009, Geyman & Maloof 2019, Hoffman & Lamothe 2019). Prave et al. (2022) also noted that although some carbonate rocks deposited in open and deeper marine environments record strongly elevated $\delta^{13}C_{carb}$ values, these are often associated with turbidite sequences and may represent the transport and redeposition of sediments from shallow water and near-shore environments, where ^{13}C enrichment occurred. However, while turbiditic sediments with very positive $\delta^{13}C_{carb}$ values have been observed in some of these ancient deep-water systems, not all occurrences of deep-water carbonate sediments document evidence of turbidity currents. Further, some formations record strongly elevated $\delta^{13}C_{carb}$ values without evidence for local water mass restriction, such as the Nash Fork Formation with $\delta^{13}C_{carb}$ up to +30‰ (Bekker et al. 2003, Planavsky et al. 2012).

The importance of local processes and a potential facies-dependent expression of the LJE has also been noted by Frauenstein et al. (2009), studying the Transvaal Supergroup of South Africa: "The pattern of δ^{13}C values is clearly bimodal... The intercalation of ^{13}C-enriched (+10‰) and 'normal marine' (∼0‰) samples between and within formations, and between and within specific horizons, suggests that we are not dealing with a global signal that reflects coeval seawater DIC, but with phenomena of basinal or regional extent. Such an interpretation implies a cautionary note on application of carbon isotope stratigraphy as a global correlation tool or on interpretation of δ^{13}C anomalies as products of global redox balance for the ocean/atm system" (p. 158).

A facies-dependent expression of the LJE is supported by the occurrence of very positive $\delta^{13}C_{carb}$ in both DIC and carbonate minerals in some modern environments, including those associated with methanogenesis as discussed above. Studies of a small, highly evaporitic lake in Sinai have shown the occurrence of $\delta^{13}C_{carb}$ up to +6‰ due to high local primary productivity resulting in preferential removal of ^{12}C into organic carbon (Aharon et al. 1977, Schidlowski et al. 1985). Studies of modern sediments in the Great Bahamas Bank have also indicated that there is widespread precipitation of carbonate minerals with $\delta^{13}C$ values of approximately +5‰ largely due to ^{13}C enrichment in the DIC reservoir by photosynthesis (Swart et al. 2009, Geyman & Maloof 2019). Overall, these studies illustrate that geochemical conditions resulting in strong ^{13}C enrichments analogous to the LJE are found in the modern/recent Earth system in various shallow water environments and range in size from small, isolated niche environments to large carbonate platforms. It is possible that $\delta^{13}C_{carb}$ can vary significantly due to a consortium of local factors, and such mechanisms should be considered for the LJE, although they must produce a globally preserved ^{13}C-rich excursion that is unmatched through geologic history. Whether such factors solely operate at local scales or are instead local expressions of shifts in global conditions remains challenging to unravel.

2.5. Summary of Models

While a broad range of models have been proposed for the LJE, it remains challenging to understand this event from a uniformitarian perspective. If the LJE was globally synchronous and representative of seawater DIC (i.e., the canonical model), the magnitude and duration are difficult to reconcile with isotopic mass balance and current understandings of the carbon cycle/nutrient fluxes/f_{org}. Conversely, other models (e.g., methanogenesis/authigenic carbonate formation) can lack geological evidence and/or rely on modern analogs of very limited size that are difficult to extrapolate to a global scale. Geochronological constraints also play a key role in these models: If the LJE was not globally synchronous, then some models can be ruled out (e.g., the canonical model), and issues associated with isotope mass balance and scaling of small environmental analogs to larger systems can be alleviated.

3. COMPILATION OF DATA FROM LITERATURE

To better explore the wide range of hypotheses for the LJE, we compiled the temporal and depositional constraints on globally located LJE-bearing successions. In total 9,410 samples were compiled from published and unpublished sources, building a data set that includes measured $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$, age constraints, metamorphic grade, and depositional environment, as available. These data reveal several important trends and correlations that are used to inform our understanding of different models for the LJE.

3.1. Late Diagenesis and Metamorphism

Owing to the age of LJE carbonate successions, many samples have undergone a complex burial history. In some instances, close association with calcium sulfate evaporite deposits (i.e., gypsum/anhydrite) or intense postdepositional fluid flow has led to dedolomitization where fluids with low Mg/Ca (e.g., from dissolution of calcium sulfate minerals) and low $\delta^{13}C_{carb}$ (e.g., from oxidation of organic matter or methane) result in the replacement of dolomite by calcite (Črne et al. 2014, Kreitsmann et al. 2019); resulting rocks typically have much lower or negative $\delta^{13}C_{carb}$ values. Samples associated with the formation of significant amounts of dedolomite or secondary calcite are excluded from subsequent discussion, as they do not reflect Earth's surface environment or processes occurring during early diagenesis (see the sidebar titled Early Diagenesis).

A histogram of the $\delta^{13}C_{carb}$ from LJE successions that are subgreenschist (N = 1,843) exhibits a large peak near 0‰, with a small, broad peak ranging from approximately +5 to +8‰

EARLY DIAGENESIS

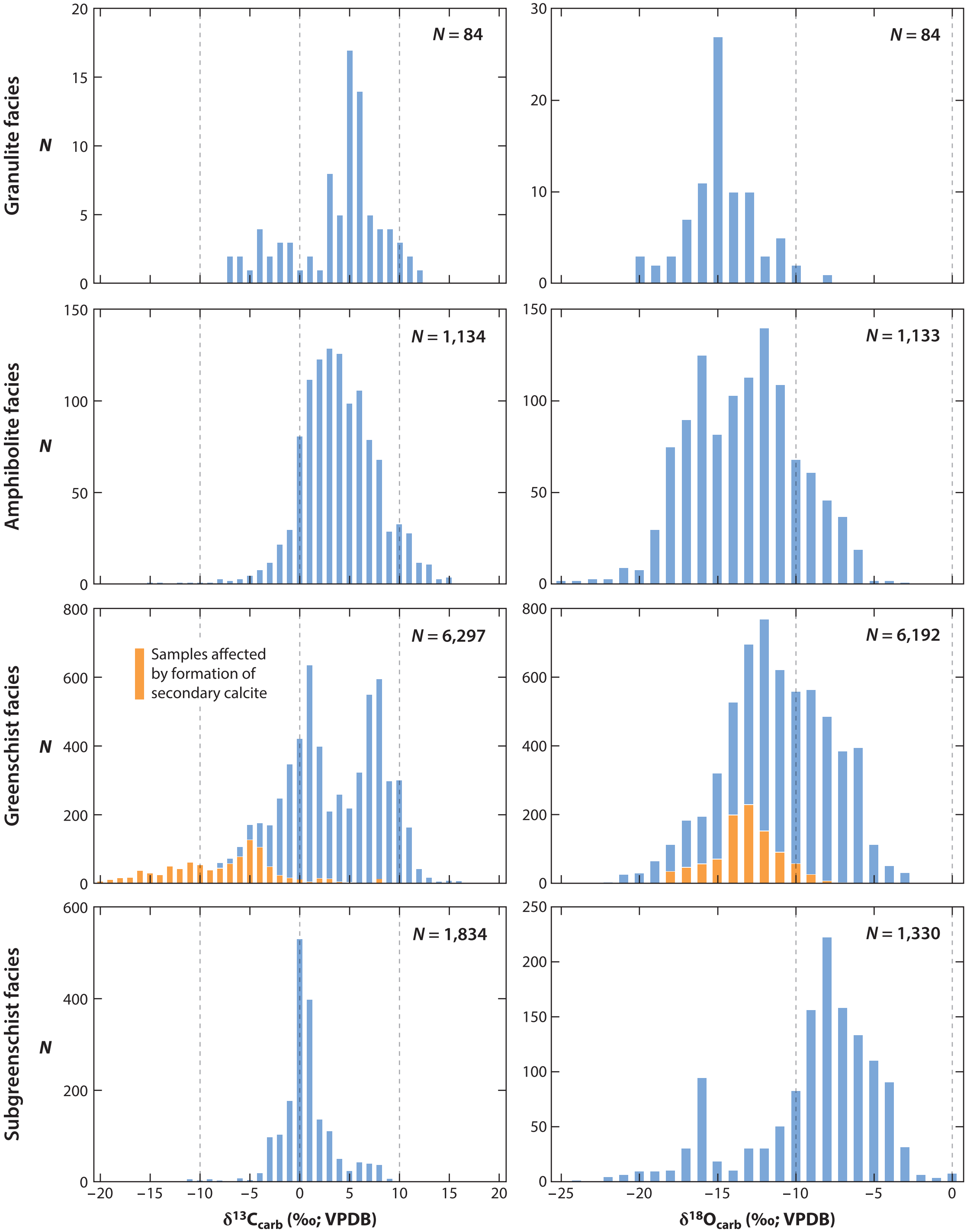
Most carbonate sediments are initially close in their geochemical composition to the seawater or local waters from which they precipitate. Local waters can be chemically distinct from the open ocean (e.g., through intense primary productivity, evaporation, or mixing with meteoric waters). During postdepositional alteration (i.e., early diagenesis) it is possible for sediments to have their geochemistry, including $\delta^{13}C_{carb}$, altered toward fluids within the sediments, which may be marine waters, evolved marine waters, or terrestrial waters (or mixtures thereof). In some cases, calcium and magnesium isotope ratios ($\delta^{44}Ca$ and $\delta^{26}Mg$, respectively) have been applied to unravel complex diagenetic histories in sediments (Fantle & DePaolo 2007, Fantle & Higgins 2014, Higgins et al. 2018) as well as to resolve the primary nature of large $\delta^{13}C_{carb}$ excursions that are difficult to explain within the traditional carbon isotope framework (Jones et al. 2019, Ahm et al. 2021, Crockford et al. 2021, Busch et al. 2022). If early diagenesis is invoked to explain the LJE, a mechanism is still required for the generation of strong ^{13}C enrichment within the early diagenetic fluids. Widespread pairing of $\delta^{44}Ca$, $\delta^{26}Mg$, and $\delta^{13}C_{carb}$ to Lomagundi-Jatuli Excursion sedimentary rocks may help us to better understand their early diagenetic history and the primary versus secondary nature of this carbon isotope excursion.

(**Figure 4**). Samples from the greenschist ($N = 5{,}749$) metamorphic facies record a strongly bimodal distribution of $\delta^{13}C_{carb}$ values, with peaks centered around 0 and +7‰, representing normal and LJE conditions, respectively. In contrast, samples from amphibolite-grade successions ($N = 1{,}078$) form a positively skewed normal distribution with a peak near +3‰. Finally, samples from granulite grade successions ($N = 84$) record a single peak near +5‰. Increasing metamorphic grade can result in dedolomitization through the replacement of dolomite by calcite and talc, during which degassing of ^{13}C- and ^{18}O-rich CO_2 drives residual carbonate minerals $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ toward lower values (Valley 1986, Črne et al. 2014). The absence of a pronounced +7‰ peak in $\delta^{13}C_{carb}$ of amphibolite facies samples compared to greenschist samples is consistent with the devolatilization that is expected, as is the overall trend toward lower $\delta^{18}O_{carb}$ values (e.g., medians of −8.0 and −14.7‰ for subgreenschist and granulite facies, respectively). Kolmogorov-Smirnov tests comparing $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ between different metamorphic facies confirm statistically significant differences, with p values consistently much less than 0.05 (i.e., confidence level much greater than 95%).

To minimize the effect of metamorphism on the data set, amphibolite and granulite grade samples are excluded from subsequent discussion, focusing instead on samples that are subgreenschist or greenschist facies. Including just these metamorphic grades, and additionally excluding samples that contain significant amounts of secondary calcite from dedolomitization, results in 7,031 samples. The filtered data show that the $\delta^{13}C_{carb}$ of baseline Paleoproterozoic carbonate rocks is most commonly 0‰, with a $\delta^{18}O_{carb}$ of −8‰. In contrast, the $\delta^{13}C_{carb}$ of LJE carbonates is +7‰, with a $\delta^{18}O_{carb}$ of −12‰. The results are strongly bimodal, with clearly defined LJE and non-LJE populations (**Figure 5**).

3.2. Depositional Environments

Compiled data were binned into three categories of depositional environment: (*a*) sabkha and peritidal; (*b*) platform and mid-upper ramp; and (*c*) lower ramp, platform slope, and deep basin. Only data with age constraints that permit deposition between 2306 ± 9 Ma to 2057 ± 1 Ma [the maximum interval for a synchronous LJE (Martin et al. 2013)] are considered, to avoid creating an artificially large population of $\delta^{13}C_{carb}$ data clustered around 0‰. Samples from all depositional



(Caption appears on following page)

Figure 4 (*Figure appears on preceding page*)

Histograms of $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ by metamorphic facies. $\delta^{13}C_{carb}$ data from Lomagundi-Jatuli Excursion (LJE) successions show clear bimodality in the greenschist facies, compared to amphibolite and granulite, suggesting metamorphism altered $\delta^{13}C_{carb}$ values in the higher grades. Subgreenschist facies samples record only a minor population of ^{13}C-enriched LJE values. $\delta^{18}O_{carb}$ data shift toward increasingly negative values with increasing metamorphic grade.

environments record peaks around 0 to +1‰ and +7 to +8‰ (**Figure 6**), although the relative size of the ^{13}C-enriched population decreases with increasing water depth, from more than 50% of samples to less than 33% (Kolmogorov-Smirnov tests result in p values much less than 0.05). Sedimentary rocks reflecting deeper water environments influenced by turbidity currents have a median $\delta^{13}C_{carb}$ of +2.8‰, compared to +0.2‰ for environments without evidence of sediment redeposition (Kolmogorov-Smirnov test p value much less than 0.05) (**Figure 7**). The Zaonega Formation, for example, is a deep-water unit with evidence for sediment redeposition and records $\delta^{13}C_{carb}$ up to +8.9‰ (**Figure 7**). However, not all deep-water sedimentary units appear to follow this paradigm; the Silverton Formation was deposited in a deep-water environment and has no reported evidence for sediment redeposition yet records a median $\delta^{13}C_{carb}$ of +9.8‰ (**Figure 7**). Units such as this may be important targets for detailed sedimentological study.

3.3. Age Constraints

Owing to the absence of robust biostratigraphic markers and the general scarcity of dateable material (e.g., volcanic tuffs with zircons for U-Pb dating, organic-rich shales for rhenium-osmium

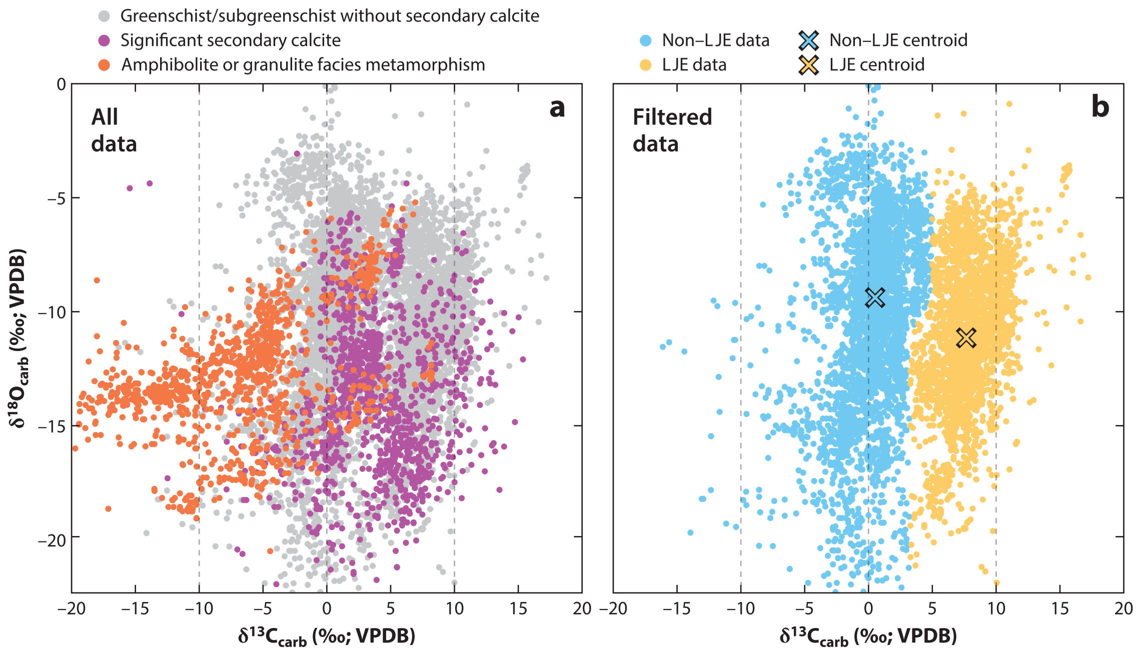


Figure 5

A cross plot of compiled $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ data from ca. 2.5 to 1.8 Ga (mostly ca. 2.3 to 2.0 Ga). (*a*) Unfiltered data showing samples containing significant secondary calcite (*purple*), affected by amphibolite or granulite facies metamorphism (*orange*), or affected by neither (*gray*). (*b*) Filtered data (*gray* in the all data panel) divided into two clusters using k-means clustering clearly reflect non-LJE versus LJE samples. The former has a centroid with $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ values of +0.4 and −9.3‰, respectively, and the latter has a centroid with $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ of +7.6 and −11.0‰, respectively.

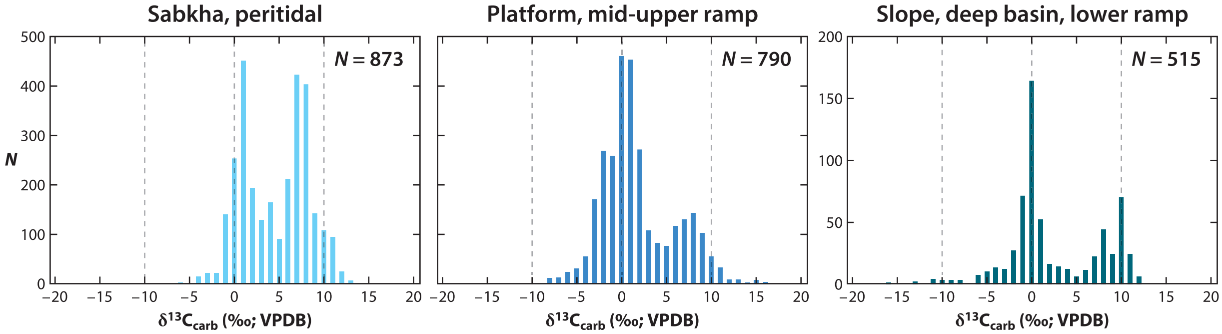


Figure 6

Histograms of $\delta^{13}C_{carb}$ from formations with age constraints that allow for deposition during the Lomagundi-Jatuli Excursion (i.e., between ca. 2300 and 2050 Ma), organized by inferred depositional environments. Peaks of approximately 0 and 7‰ are visible in all environments, although the latter population is significantly smaller in deep-water environments. The Silverton and Zaonega formations are plotted separately in **Figure 7** due to large amounts of data relative to other deep-water formations.

[Re-Os] dating, etc.), many Paleoproterozoic formations have poor age constraints; this can be further exacerbated by uncertainties on the correlation of geological units within regions (e.g., Rasmussen et al. 2019). Cumulatively, this has resulted in large uncertainties regarding the timing of the LJE, with the onset constrained between 2306 ± 9 Ma to 2221 ± 5 Ma, and the termination constrained between 2106 ± 8 Ma to 2057 ± 1 Ma (Martin et al. 2013). However, this age range is contingent on the LJE being globally synchronous, which has not been robustly demonstrated. Considering the maximum and minimum age constraints for individual formations without assuming global synchroneity indicates that many formations could have been deposited tens to

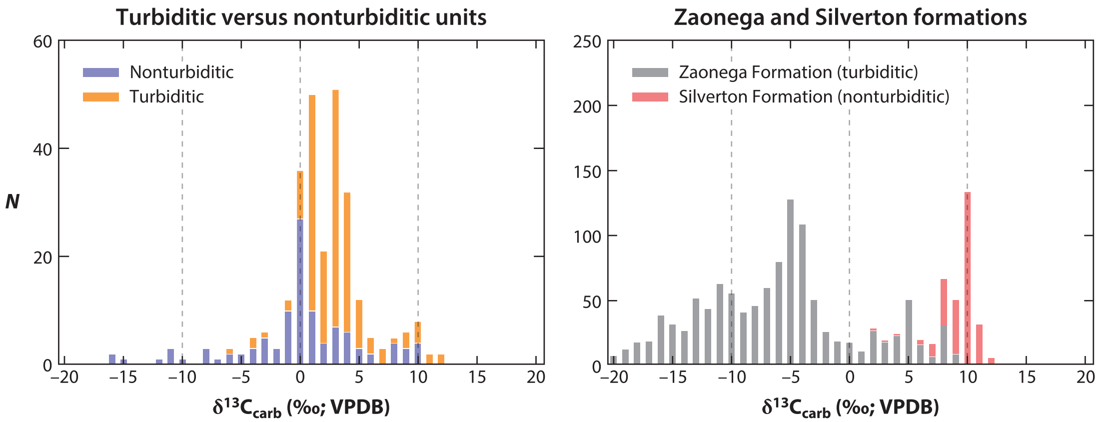


Figure 7

Histograms of $\delta^{13}C_{carb}$ from samples deposited during the Lomagundi-Jatuli Excursion (LJE) in deep-water environments. Samples deposited in turbiditic environments have a modal $\delta^{13}C_{carb}$ that is several permil higher than nonturbiditic units, potentially speaking to the importance of sediment redeposition from shallow water systems where the LJE is strongly expressed. The Silverton and Zaonega formations are plotted separately because they comprise the vast majority of these data. The Silverton Formation is anomalous for recording very positive $\delta^{13}C_{carb}$ in a deep-water environment without evidence for sediment redeposition, whereas the deep-water Zaonega Formation was deposited in a deep-water environment with evidence for turbidites and records $\delta^{13}C_{carb}$ up to +9‰. Note that the very large negative tail is likely due to the formation of secondary calcite, and as such, the Zaonega Formation has been excluded from discussion elsewhere.

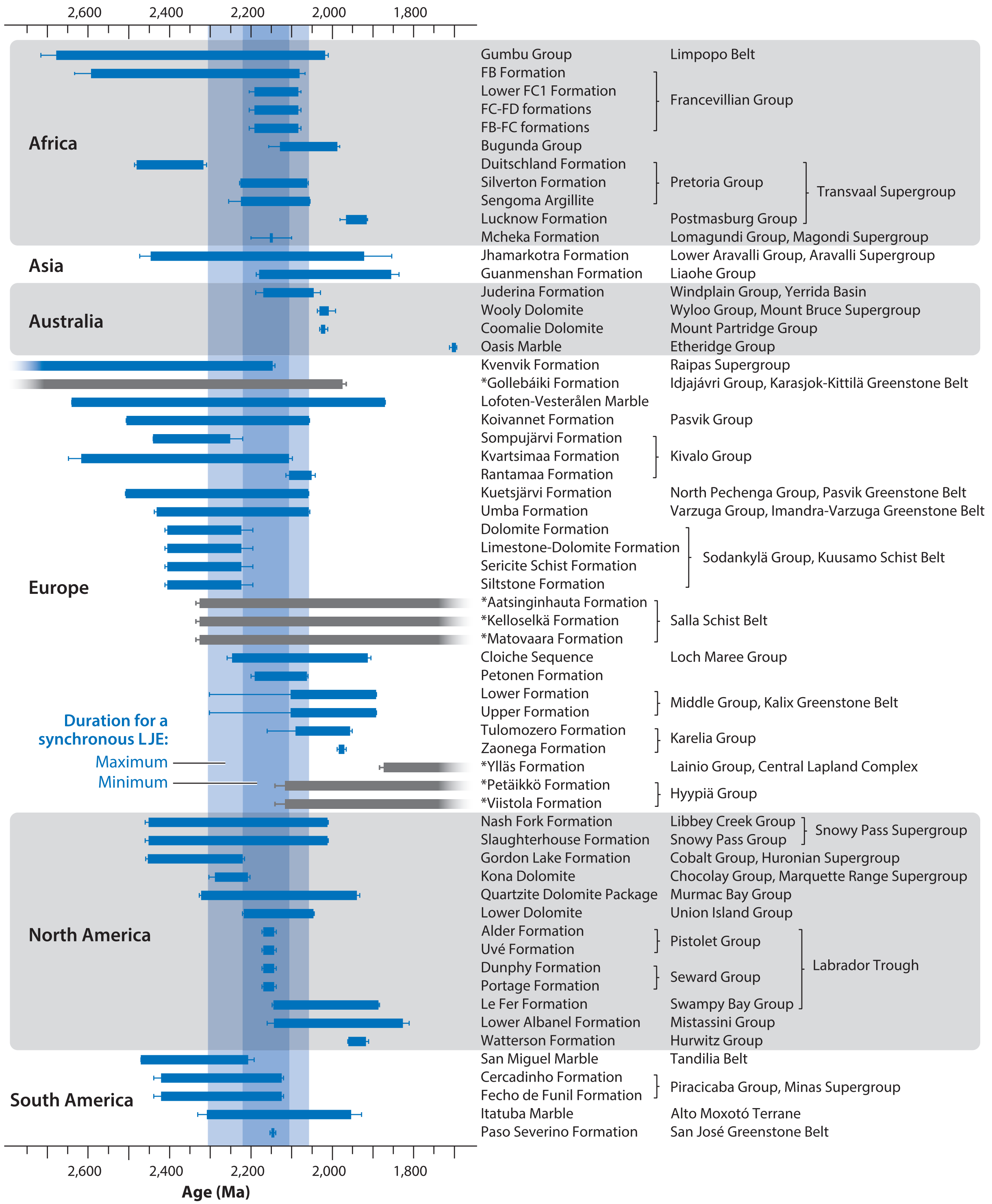
hundreds of millions of years outside of the time interval indicated above and span from more than 2.5 to less than 1.9 Ga (**Figure 8**).

4. WAS THE LOMAGUNDI-JATULI EXCURSION GLOBALLY SYNCHRONOUS?

Whether or not the LJE was globally synchronous is central to whether it should be interpreted as a consequence of mechanisms that shifted global seawater $\delta^{13}C_{DIC}$ rather than processes that shifted $\delta^{13}C_{DIC}$ within the local water or sediment column(s). Supporting the LJE being a series of asynchronous, local events are data indicating that there are several packages of Paleoproterozoic carbonate rocks with strongly positive $\delta^{13}C_{carb}$ values deposited outside the LJE time interval (ca. 2.3 to 2.05 Ga) (**Figure 8**). For example, the Wooly Dolomite in Australia was deposited between ca. 2031 ± 6 and 2008 ± 16 Ma, and it records $\delta^{13}C_{carb}$ values up to +8‰ (Bekker et al. 2016). In South Africa, the Lucknow Formation records $\delta^{13}C_{carb}$ values up to +10‰, and geochronologic data from Beukes et al. (2019) suggest it was deposited between ca. 1966 ± 15 and 1916 ± 1 Ma. A single sample from the Watterson Formation (ca. 1960 ± 2.2 to 1917 ± 6 Ma) of Nunavut, Canada, records $\delta^{13}C_{carb}$ values of approximately +8.5‰ (Hofmann & Davidson 1998). Some formations in Finland with elevated $\delta^{13}C_{carb}$ values may have been deposited outside of this interval as well, although contextual information is sparse (Karhu 1993). Given that ^{13}C-enriched carbonate sediments can be found in niche modern depositional environments (as discussed in Sections 2.3.1 and 2.4), these isolated occurrences of high $\delta^{13}C_{carb}$ alone cannot provide strong evidence against a synchronous LJE, even if they were deposited outside the time interval typically assigned to the LJE. These strata do, however, indicate that extreme ^{13}C enrichment is not exclusive to the ca. 2.3–2.05 Ga interval and that the LJE as a series of discrete, local events should be considered.

For any given unit to record a significant proportion of the 130–250-million-year duration hypothesized for a synchronous LJE, the net sediment accumulation rate of the unit would have to be much lower than in most Phanerozoic carbonate systems (**Figure 9**). If LJE strata instead record more typical (i.e., faster) net accumulation rates, then many LJE-bearing formations record a few tens of millions of years at most, or only a small portion of the interval proposed if the LJE carbon isotope excursion was synchronous. It follows from this that it is very unlikely any two LJE-bearing units are truly coeval. Instead, the geological record of the LJE may be composed of many small fragments of a single very long carbon isotope excursion, each fragment of which is itself highly incomplete (Miall 2015). Alternately, each formation may record an asynchronous excursion occurring on a basin scale, as a local manifestation of a broader shift in Earth system conditions during an LJE interval spanning hundreds of millions of years. If sedimentary rocks recording the LJE did accumulate at very slow rates, it would require either that they are unrepresentative of typical sediment accumulation rates during this time or that vast spatial areas were accumulating carbonate sediments. Alternatively, if such slow accumulation rates were a response to tectonic quiescence (Condie et al. 2009, 2022) and much lower CO_2 degassing, this undermines central assumptions within the canonical model, which assumes a constant degassing source and, by extension, a constant degassing rate. Together these observations merit caution in extrapolating global inferences based on many LJE-bearing successions.

The application of Re-Os radiometric dating of organic-rich shales may help to constrain the depositional ages of units that have historically been difficult to date, given the occurrence of organic-rich units overlying many LJE-bearing successions, and the demonstrated success of combined U-Pb zircon and Re-Os shale dating for better establishing synchroneity of global-scale events in the Precambrian (Rooney et al. 2015). Beyond understanding the LJE, this is also essential for Paleoproterozoic chemostratigraphic correlation, as many formations have been assigned



(Caption appears on following page)

Figure 8 (*Figure appears on preceding page*)

Maximum and minimum age constraints for Paleoproterozoic formations that record $\delta^{13}C_{carb}$ greater than +5‰. If the Lomagundi-Jatuli Excursion (LJE) is not assumed to be a globally synchronous carbon isotope excursion, existing age constraints suggest that LJE-like conditions could have persisted for hundreds of millions of years before or after the age interval typically assigned to the LJE (*lighter* and *darker blue vertical bars* correspond to maximum and minimum LJE intervals from Martin et al. 2013). Medium gray horizontal bars with an asterisk beside the formation name indicate units with limited contextual information.

depositional ages of ca. 2300 to 2050 Ma based on the occurrence of very positive $\delta^{13}C_{carb}$ values alone. The possibility of multiple asynchronous LJE-like excursions and therefore nonunique solutions to $\delta^{13}C_{carb}$ chemostratigraphic correlations (e.g., Hay et al. 2019) could potentially lead to incorrect correlations, with erroneous implications for reconstructing the Paleoproterozoic Earth system. As an example, drill core of Cenozoic carbonate sediments from the Great Bahamas Bank and Enewetak Atoll (western Pacific Ocean) both record pronounced $\delta^{13}C$ excursions with magnitudes of approximately 8‰ (Smith & Swart 2022). The magnitude and geometry (i.e., the overall profile of the $\delta^{13}C_{carb}$ curve) of the carbon isotope excursions in each of these sites are remarkably similar, and both regions record geochemical evidence that indicates these excursions were created by meteoric diagenesis during relative sea level fall. Due to the high-resolution geochronological constraints possible in the Quaternary, it was determined that these two excursions occurred

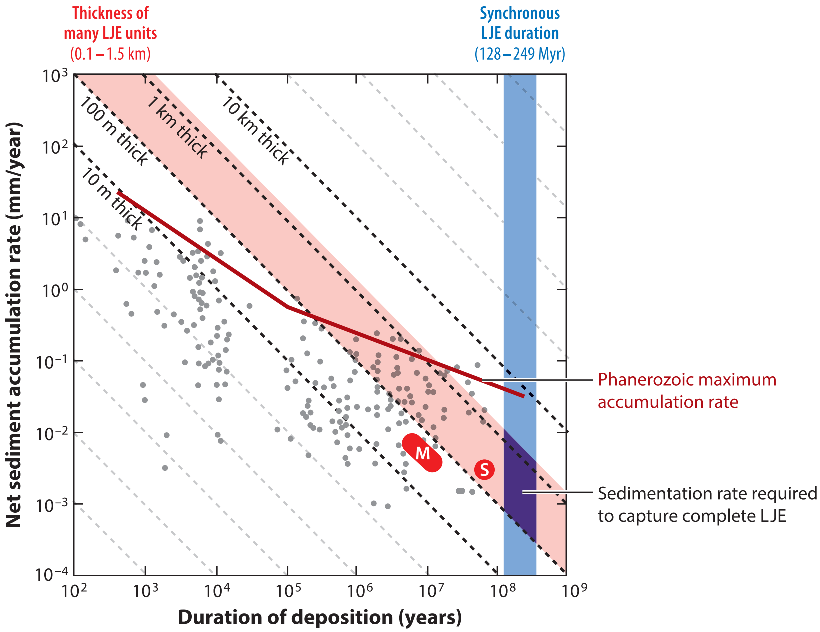


Figure 9

A comparison of sedimentation rates, unit thicknesses, and the Lomagundi-Jatuli Excursion (LJE) timescale. The depositional rates required for LJE carbonates to capture a significant amount of the LJE in a synchronous model (*purple field*) require implausibly low sedimentation rates. Diagonal dashed lines connecting the net sediment accumulation rate (*y*-axis; logarithmic) and time (*x*-axis; logarithmic) represent sediment package thickness. Calculated LJE net sediment accumulation rates would be below almost all observed Phanerozoic carbonate systems (*gray dots*) (Schlager 2000) and comparable to or below the Marinoan (M) and Sturtian (S) glacial diamictites (in *red*) (Partin & Sadler 2016), during which sediment deposition effectively ceased. If sediment accumulation rates were comparable to Phanerozoic systems, LJE carbonate units would generally record less than a few tens of millions of years.

ca. 2.0–0.2 and 5.5–2.5 Ma, respectively, and can therefore be resolved as asynchronous. Although these were negative carbon isotope excursions, they offer a cautionary tale that in the absence of a robust geochronological framework (typical for Paleoproterozoic successions), significant changes in $\delta^{13}C_{carb}$ chemostratigraphic curves driven by diagenetic mechanisms may be remarkably difficult to distinguish as globally synchronous or asynchronous. Further, geochronologically resolving two such events from Paleoproterozoic sedimentary rocks would be very challenging given existing analytical uncertainties (even if the dateable material itself was not rare). Taken as a whole, it appears that unambiguous evidence for the LJE as a globally synchronous carbon isotope excursion representative of seawater DIC is lacking. Geochronological constraints are sufficiently sparse that global synchroneity cannot objectively be assumed, and at least several formations containing comparable $\delta^{13}C_{carb}$ trends appear to have been deposited tens of millions of years after the LJE time interval.

5. ARE LOMAGUNDI-JATULI EXCURSION CARBONATE ROCKS RECORDING SEAWATER DISSOLVED INORGANIC CARBON?

In addition to considering the synchroneity of the LJE, it is essential to consider whether the LJE is representative of seawater $\delta^{13}C_{DIC}$. If this is the case, the sedimentary record should document complete rising and falling limbs showing a globally synchronous transition from $\delta^{13}C_{carb}$ values of approximately 0‰ to those of greater than +5‰, and back to 0‰ (**Figure 3**), analogous to other carbon isotope excursions observed in the geological record. However, no studies have identified complete rising and/or falling limbs within a stable depositional setting; documentation of any rising limbs is particularly sparse (Bekker 2015). The Proterozoic ocean likely had a larger DIC reservoir than the modern ocean (Bartley & Kah 2004, Cantine et al. 2020), thus making a geologically instantaneous shift of seawater $\delta^{13}C_{DIC}$ unlikely and the absence of rising and falling limbs even more disconcerting.

Could the LJE have been many local excursions in water column $\delta^{13}C_{DIC}$, which occurred either synchronously or asynchronously, while open ocean $\delta^{13}C_{DIC}$ remained near 0‰? Data compiled here and by Prave et al. (2022) indicate that positive $\delta^{13}C_{carb}$ values characteristic of the LJE are most pronounced in shallower, more restricted settings, pointing toward processes operating at a local rather than global scale. Although the specific mechanism that could have caused these ^{13}C enrichments uniquely in shallow water environments at this point in Earth history remains uncertain, the possibility does ameliorate some issues with a global shift in $\delta^{13}C_{DIC}$. First, limiting very positive $\delta^{13}C_{carb}$ values to shallow/restricted environments would allow for asynchronous, local ^{13}C-enrichment excursions. Second, a facies-dependent expression of $\delta^{13}C_{carb}$ may help to explain the large range in values from approximately +5 to +15‰ (and locally up to +30‰) observed in LJE-carbonate rocks from global locations. Finally, maintaining isotopic mass balance does not require a complete reimagining of the global carbon cycle if extreme ^{13}C enrichment was limited to shallow water and restricted regions, much like in certain modern environments, rather than the entire seawater DIC reservoir.

It is interesting to note that in many regions, the LJE terminates with the deposition of organic-rich shales. This is the case in Fennoscandia (Zaonega Formation), Gabon (FB, FC, and FD formations), the Union Island Group (Black Shale Unit), Wyoming (M3 Member of the Nash Fork Formation), and elsewhere. The timing of these organic-rich sedimentary rocks is seemingly late, given that—all else being equal—they should be coeval with the ^{13}C-enriched sedimentary carbonates, rather than postdating them. An alternate explanation could be that these organic-rich sedimentary rocks reflect the drowning of carbonate environments (e.g., Bekker et al. 2003), and therefore termination of the restricted and shallow water environments where the carbon isotope excursion of the LJE was most strongly expressed.

6. TOWARD AN INTEGRATED UNDERSTANDING OF THE LOMAGUNDI-JATULI EXCURSION/GREAT OXIDATION EVENT EARTH SYSTEM

Based on existing geochronological evidence and the relationship between depositional environments and $\delta^{13}C_{carb}$, it seems the LJE may represent an interval where Earth's surface conditions allowed for the expansion of unusual (bio)geochemical conditions in shallow and restricted settings, rather than a wholesale shift within the global carbon cycle. The possibility that the LJE was not a globally synchronous carbon isotope excursion in the traditional sense of shifting seawater $\delta^{13}C_{DIC}$ is compelling: The most positive $\delta^{13}C_{carb}$ values are observed in shallow and restricted environments, and many deep environments with elevated $\delta^{13}C_{carb}$ values show evidence for redeposition of sediments likely sourced from shallower environments. The absence of an unambiguous onset/termination of the LJE (predicted if it was representative of seawater $\delta^{13}C_{DIC}$) also supports this hypothesis. A facies-dependent expression of the LJE also avoids a complete reorganization of the global carbon cycle and redox budget. The possibility that the LJE was not reflective of global ocean DIC in no way diminishes its uniqueness: The large, very positive $\delta^{13}C_{carb}$ population deposited across many regions and environments over this interval is still remarkable and unique in Earth history.

If the LJE was a series of local excursions in $\delta^{13}C_{DIC}$, we speculate that it may have occurred during the subaerial emergence of continents and resulting widespread creation of (semi)restricted shallow basins. The water column $\delta^{13}C_{DIC}$ within individual basins may have been elevated through enhanced primary productivity driven by increased nutrient flux (e.g., phosphate) during continental emergence and chemical weathering under a high-CO_2 atmosphere, and potentially water-atmosphere CO_2 disequilibrium (Beeler et al. 2020). Sulfur isotope data indicate transient oxygenation in the lead up to the LJE (Poulton et al. 2021), which may have also increased nutrient fluxes through oxidative weathering. Phylogenomic analyses suggest that photosynthetic eukaryotes originated in freshwater environments during the mid-Paleoproterozoic (Sánchez-Baracaldo et al. 2017), and perhaps the emergence of continents and development of semirestricted basins provided environments for diversification and expansion. The local expression of the LJE may have ended when basins flooded due to either local or global tectonic events, and/or the global mechanisms driving the LJE interval (i.e., the global factors allowing for local expression of very positive $\delta^{13}C_{carb}$) tapered off as stable continents were established and nutrient fluxes returned to baseline conditions.

Nonetheless, aspects of the canonical interpretation for the LJE remain compelling. Perhaps most importantly, the LJE and GOE broadly occur around the same time, thereby temporally linking the oxidation of Earth's surface environment and a large carbon isotope excursion (Karhu & Holland 1996, Bekker & Holland 2012, Poulton et al. 2021). This offers a self-consistent and coherent explanation that is seemingly supported by a range of redox proxies (e.g., Partin et al. 2013, Hardisty et al. 2014, Kipp et al. 2017) and evidence for elevated primary productivity inferred during the LJE (Crockford et al. 2019, Hodgskiss et al. 2019a). At the same time, models that attempt to understand changes in redox chemistry across this interval by assuming relative timing based on $\delta^{13}C_{carb}$ (e.g., Kipp et al. 2017, Hodgskiss et al. 2021) run the risk of associating two potentially separate events, neither of which is well understood in terms of timing and underlying mechanisms. Also worth considering is whether the LJE may be related to the proposed Paleoproterozoic Snowball glaciations (or events that occurred in their aftermath), although evaluation of cause-effect relationships is difficult due to uncertainties of timing and how many of these glacial events occurred (Gumsley et al. 2017).

Rectifying the range of viewpoints on the LJE is essential for understanding the critical biological and geochemical changes occurring across Earth's surface environments during this pivotal time. Central to achieving this aim will be international collaborative efforts (e.g., FAR-DEEP) designed to extract maximum sedimentological, geochemical, and geochronological data from complete successions, ideally using drill cores collected specifically for research purposes. This approach would minimize sampling gaps, create detailed reconstructions of depositional environments, and allow for intensive and systematic collection of geochronological data to create a robust geological framework. Ultimately, this would allow for the application of geochemical proxies to better understand the cause-and-effect relationships between the GOE and LJE, and their driving mechanisms. Other important progress can be made on the fronts of geochronology, high-resolution $\delta^{13}C_{carb}$ chemostratigraphy, and sedimentology (see the section titled Future Issues).

More broadly, the significant uncertainties associated with the LJE highlighted here urge caution on the use of $\delta^{13}C_{carb}$ chemostratigraphy for poorly understood and time-calibrated carbon isotope excursions in the Precambrian, particularly when compared to Phanerozoic carbon isotope excursions. The incorrect application of these tools could result in the interpretation of spatially and temporally isolated changes in $\delta^{13}C_{carb}$ in terms of a global carbon isotope mass balance (Ahm & Husson 2022), possibly leading to erroneous conclusions regarding global significance and synchroneity. Continued work to resolve the chronology and local versus global nature of these $\delta^{13}C_{carb}$ shifts is therefore desperately needed as researchers work to better understand the evolution of Earth's carbon cycle.

The LJE is often taken as a robust foundation upon which biological and environmental interpretations are built. Although these strongly ^{13}C-enriched carbonate rocks are seemingly related to, and often occur in association with, rocks that record evidence of atmospheric oxygenation, the cause-effect relationships remain unclear and the underlying mechanisms strongly debated. Despite more than 50 years of research, possible explanations for the LJE are diverging rather than converging, and it remains equivocal whether the LJE was synchronous, reflective of processes occurring within the open ocean, local water column, or sediment column, or linked to oxygenation of the atmosphere/oceans. In many ways, this speaks to the remarkable and exciting challenge of understanding the LJE, the creative and data-driven approaches needed to understand an event that challenges our understanding of Earth history and the global carbon cycle, and the limits of what is interpretable from ancient sedimentary archives.

FUTURE ISSUES

1. The timing of the Lomagundi-Jatuli Excursion (LJE) is still highly uncertain. Improved geochronological age constraints would clarify the relationship between the Great Oxidation Event (GOE) and LJE, and test whether the LJE was synchronous or asynchronous. This also has important implications for the global correlation of strata.
2. The $\delta^{13}C_{carb}$ shifts marking the beginning/end of the LJE are poorly constrained in the geological record. However, the size of the seawater dissolved inorganic carbon (DIC) reservoir requires that such a shift should be marked by complete rising and falling limbs as a new equilibrium is reached. Documentation of this transition is lacking and may inform whether the LJE was reflective of seawater DIC or more restricted settings such as the local water/sediment column(s).
3. Many LJE carbonate rocks are directly overlain by organic-rich black shales. The application of rhenium-osmium geochronology may help to test whether these represent widespread (i.e., synchronous) or local (i.e., asynchronous) flooding events and therefore

help us to understand synchroneity of the end of the LJE. The application of shale redox proxies may help us to understand changes in seawater chemistry during this time, and whether the end of the LJE was linked to a shift toward more reducing conditions.

4. The LJE and GOE have been explicitly linked in some models, predicting that a shift toward strongly positive $\delta^{13}C_{carb}$ should coincide with a shift toward more oxic conditions. The integration of carbonate-based redox proxies [e.g., I/(Ca+Mg) ratios, Ce anomalies, and Mo/U isotope ratios] and $\delta^{13}C_{carb}$ chemostratigraphy may therefore help us to understand the cause-effect relationship between these two events. Application of rare earth elements and yttrium systematics may help us to determine whether there is a relationship between $\delta^{13}C_{carb}$ and the degree of restriction/terrestrial versus marine influences.
5. A facies-dependent expression of the LJE predicts that open ocean $\delta^{13}C_{DIC}$ values remain near 0‰, while the $\delta^{13}C_{carb}$ of sediments deposited in shallow and restricted environments shifts to strongly positive values. In this model, the occurrence of very positive $\delta^{13}C_{carb}$ in deep-water environments is the result of sediment transport from shallow water environments. Detailed sedimentological studies of deep-water units with strongly positive $\delta^{13}C_{carb}$ values would provide a critical test of this hypothesis.
6. Investigation of authigenic carbonate rocks has been carried out in sedimentary rocks from the latter half of the Neoproterozoic, but systematic study has not been carried out during the LJE interval. This will test whether authigenic carbonate formation was an important sink of ^{12}C during the LJE.
7. Changes in the $\delta^{13}C$ of carbonate sediments during early diagenesis have been invoked to explain other enigmatic carbon isotope excursions in Earth history. The integration of calcium- and magnesium-stable isotope ratio data with traditional $\delta^{13}C_{carb}$ for LJE carbonate rocks may help to untangle the nature of early diagenesis and the degree to which the LJE records a primary environmental signal.

DISCLOSURE STATEMENT

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