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## RESEARCH ARTICLE

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### Special Section:

Illuminating a Warmer World:  
Insights from the Paleogene

### Key Points:

- Combined carbonate isotope approach disentangles paleoclimate/diagenesis despite complex environmental histories on Ellesmere/Axel Heiberg
- Carbon and oxygen isotope values highlight hydrologic seasonality in the Arctic during the Paleogene
- Temperatures from carbonate clumped isotopes are consistent with vitrinite reflectance burial temperature, suggesting variable deformation

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## Paleogene Paleohydrology of Ellesmere and Axel Heiberg Islands (Arctic Canada) From Palustrine Carbonates

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**Abstract** Ancient greenhouse periods are useful analogs for predicting effects of anthropogenic climate change on regional and global temperature and precipitation patterns. A paucity of terrestrial data from polar regions during warm episodes challenges our understanding of polar climate responses to natural/anthropogenic change and therefore our ability to predict future changes in precipitation. Ellesmere and Axel Heiberg Islands in the Canadian Arctic preserve terrestrial deposits spanning the late Paleocene to middle Eocene (59–45 Ma). Here we expand on existing regional sedimentology and paleontology through the addition of stable ( $\delta^{13}\text{C}$ ,  $\delta^{18}\text{O}$ ) and clumped ( $\Delta_{47}$ ) isotope analyses on palustrine carbonates.  $\delta^{13}\text{C}$  isotope values range from  $-4.6$  to  $+12.3\text{‰}$  (VPDB), and  $\delta^{18}\text{O}$  isotope values range from  $-23.1$  to  $-15.2\text{‰}$  (VPDB). Both carbon and oxygen isotope averages decrease with increasing diagenetic alteration. Unusually enriched carbon isotope ( $\delta^{13}\text{C}$ ) values suggest that analyzed carbonates experienced repeated dissolution-precipitation enrichment cycles, potentially caused by seasonal fluctuations in water availability resulting in summer carbonate dissolution followed by winter carbonate re-precipitation. Stable isotopes suggest some degree of precipitation seasonality or reduction in winter water availability in the Canadian Arctic during the Paleogene. Clumped ( $\Delta_{47}$ ) temperature estimates range from 52 to 121°C and indicate low temperature solid-state reordering of micritic samples and diagenetic recrystallization in sparry samples. Average temperatures agree with vitrinite reflectance data for Eureka Sound Group and underlying sediments, highlighting structural complexity across the region. Broadly, combined stable and clumped isotope data from carbonates in complex systems are effective for describing both paleoclimatic and post-burial conditions.

**Plain Language Summary** Paleogene paleohydrology in Arctic Canada has been a question of much debate in recent years, with competing ideas about conditions characterized as either “polar monsoon” or “ever-wet” regimes. Proxy records from Ellesmere and Axel Heiberg islands have been central to this debate, but complex stratigraphy and burial histories have hampered a resolution. Here we employ a combined carbonate isotope ( $\delta^{13}\text{C}$ ,  $\delta^{18}\text{O}$ ,  $\Delta_{47}$ ) approach to disentangle paleoclimate and diagenetic signals. Using palustrine carbonates (which host previously evaluated paleobotanical remains) within a new stratigraphic framework, we demonstrate that  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values from primary-fabric samples indicate a distinct, but non-monsoonal hydrologic seasonality during the late Paleocene/early Eocene. We also demonstrate that  $\Delta_{47}$  temperatures reflect both shallow burial reordering and more intense remineralization, which are consistent with vitrinite reflectance studies showing variable deformation and geothermal gradients in the Sverdrup Basin. This combined isotopic approach is novel in the region, and can be applied to other regions of interest in deep time.

## 1. Introduction

Predicting future changes to global precipitation patterns is critical for global food security, water resource management, and extreme event preparedness (IPCC, 2021). However, current precipitation predictions vary significantly among climate models due to their distinct parameters (e.g., RCP Scenarios; IPCC, 2021; USGCRP, 2018). A better understanding of precipitation dynamics during past greenhouse periods is required to improve future precipitation simulations (QDRR, 2010; Tierney et al., 2020). As the polar regions are particularly responsive to global climate change, improvement to our overall understanding of past high latitude hydrological

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regimes contributes to models of both regional and global paleohydrology and to our capacity to predict anthropogenic change.

Proxy records from past greenhouse periods exist for some regions and time periods; however, the majority of climate records currently available from Mesozoic and Paleogene warm periods are from marine environments, which may temporally lag corresponding terrestrial events (Mix & Ruddiman, 1984), and typically show diminished responses to warming (Diffenbaugh & Field, 2013). Modern climate monitoring and simulations of future warming have also shown that high-latitude regions respond dramatically to rapid warming events and can further influence the global climate (USGCRP, 2018), highlighting the need to increase our understanding of high-latitude climate dynamics. Some terrestrial proxy records from greenhouse periods exist for the Arctic (e.g., Greenwood et al., 2010; Guemas et al., 2013; Salpin et al., 2019; Suan et al., 2017; Sunderlin et al., 2011; West et al., 2015, 2020; Willard et al., 2019; Zimov et al., 2006) and southern high latitudes (e.g., Carpenter et al., 2012; Contreras et al., 2013; Francis & Poole, 2002; Ivany et al., 2011; Jacques et al., 2014; Pross et al., 2012; Reguero et al., 2002). Nevertheless, the amount of data available for terrestrial areas is comparatively low and often complicated for these crucial regions (e.g., Huber & Caballero, 2011); therefore, expanding our inventory of high-latitude terrestrial proxy records will continue to improve our understanding of the function of the hydrologic cycle during past greenhouse periods (e.g., Carmichael et al., 2016).

Accessible land area in polar regions is limited and deep-time archives are often obscured by subsequent deformation, making it difficult to unravel environmental histories. In particular, the Canadian Arctic has experienced tectonic reorganization since the Mesozoic, including significant burial and exhumation of certain terranes (e.g., Ricketts & Stephenson, 1994; Embry & Beauchamp, 2008; von Gosen et al., 2019). Previous studies found extension and rapid subsidence of the eastern part of the region (Sverdrup Basin), which resulted in increased sedimentation between the Late Cretaceous and the middle Eocene. This period of deposition was followed by regional uplift and denudation from the middle Eocene onward (Figure 1; e.g., Dewing & Obermajer, 2011; Dewing & Sanei, 2009; Embry & Beauchamp, 2008; von Gosen et al., 2019). Understanding such complex sedimentary basin dynamics can be important for interpreting samples within the context of their tectonic histories and evaluating robustness of paleoclimate records.

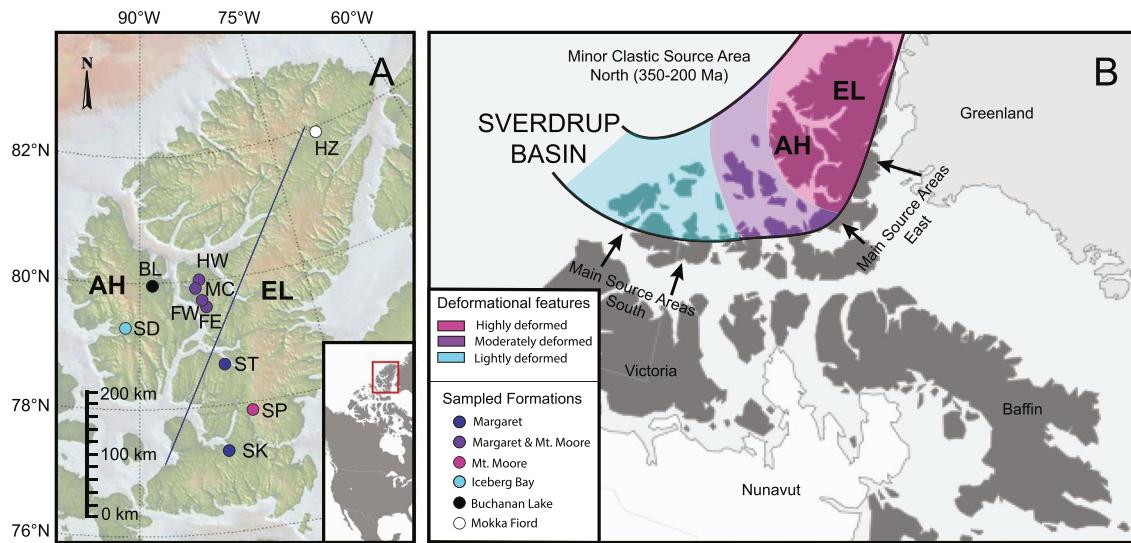
One way to address both climate and tectonic evolution questions in complex settings like the Canadian Arctic Archipelago is to employ a combination of traditional stable and novel clumped isotope systems to identify pre- and post-burial influences on carbonate rocks, and to thereby separate paleoclimatic from burial/deformation signals (e.g., Parrish et al., 2018; Winkelstern & Lohmann, 2016). The main objectives of this study are to use the carbon, oxygen, and clumped isotopic compositions of terrestrial carbonates to determine (a) if the Paleogene Arctic hydrologic system experienced some form of seasonality or periodicity in water availability, and (b) if paleoclimate information can be disentangled from signals generated by burial and subsequent carbonate alteration using a multi-proxy approach. Specifically, we analyze both the (relatively burial-insensitive) stable and (relatively burial-sensitive) clumped isotope compositions of palustrine carbonates from Ellesmere and Axel Heiberg islands (Nunavut, Canada) and use these complex data sets to provide new constraints on both Arctic paleohydrology and burial/deformation in the eastern Sverdrup Basin.

## 2. Background

### 2.1. Paleogene Arctic Climate

The modern Canadian Arctic environment is a polar desert, and is typified by a cold, dry climate inhabited by cold-adapted flora and fauna. However, during the early Paleogene, the Canadian Arctic was home to forests dominated by temperate deciduous hardwoods, conifers, and ferns (McIver & Basinger, 1999; West et al., 2019), and a diverse polar fauna consisting of thermophilic animals such as alligators, snakes, turtles, and tapirs (Eberle et al., 2014; Eberle & Greenwood, 2012; Estes & Hutchison, 1980).

The Canadian Arctic region during the early Paleogene was broadly temperate and wet (Eldrett et al., 2014; Greenwood et al., 2010; West et al., 2020), and previous paleobotanical studies have indicated these regions experienced high mean annual precipitation (>>150 cm/year), mesothermal conditions (mean annual temperature (MAT) ~12–15°C), and moderate winter temperatures (Cold month mean temperature (CMMT) > 0°C) (West et al., 2015, 2020). The forests of the Canadian Arctic grew during a warm greenhouse interval when global temperatures were much higher than modern, and the global climate system was episodically punctuated by short-lived transient hyperthermal events (McInerney & Wing, 2011; Sluijs et al., 2009; Zachos et al., 2008).



**Figure 1.** Maps of Ellesmere (EL) and Axel Heiberg (AH) islands. (a) Topographic map for Ellesmere (EL) and Axel Heiberg (AH) islands. Axel Heiberg Island contains the sample locations: Buchanan Lake (BL) and Strand Fiord (SD). Ellesmere Island contains the sample locations: Lake Hazen (HZ), Hot Weather Creek (HW), Mosquito Creek (MC), Fosheim Anticline West (FW), Fosheim Anticline East (FE), Strathcona Fiord (ST), Split Lake (SP), and Stenkul Fiord (SK). Topographic data from GeoMapApp (Ryan et al., 2009). Blue line in Panel A indicates NE-SW transect shown in Figure 6. (b) Placement of the Sverdrup Basin across the Canadian Arctic Archipelago, with sediment source areas indicated. Deformation regimes in the Sverdrup Basin include “highly deformed” with thrust faults and significant folding; “moderately deformed” with folding and salt domes; and “lightly deformed” where there is very mild deformation with broad folds (e.g., Embry & Beauchamp, 2008).

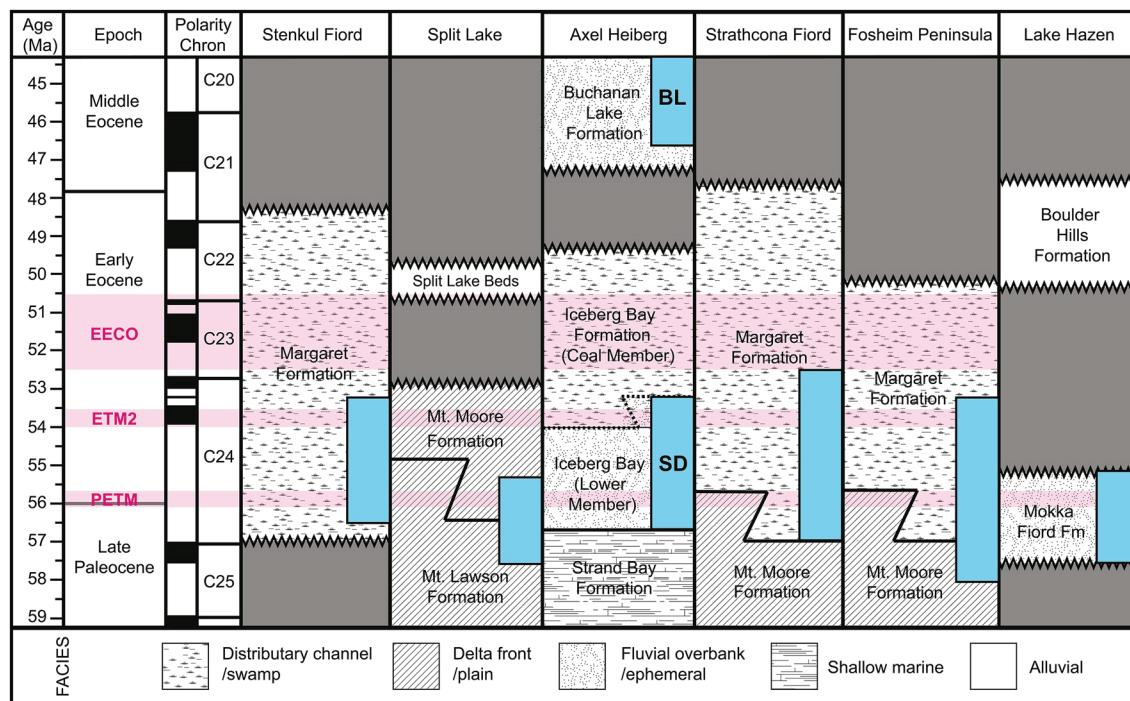
These intense episodes of global warming are considered to be some of the most abrupt and dramatic climatic warming events of the entire Cenozoic and represent some of the warmest intervals of the last 66 million years (McInerney & Wing, 2011; Westerhold et al., 2020; Zachos et al., 2008). Perhaps the best studied and most dramatic of these events is the Paleocene-Eocene Thermal Maximum (PETM), which was characterized by a large negative carbon isotope excursion ( $>4\text{\textperthousand}$  in terrestrial archives) and a subsequent global temperature increase of approximately 5–8°C in  $\leq 20\text{ky}$  (e.g., Eldrett et al., 2014; Sluijs et al., 2009; Zachos et al., 2008).

The PETM and other Paleogene warming events not only increased mean global temperatures, but also affected the global hydrological system, including precipitation intensity and distribution (Hyland & Sheldon, 2013; Hyland et al., 2017; Krishnan et al., 2014; Pagani et al., 2006; Westerhold et al., 2020; Zachos et al., 2008). The hydrological response on a regional scale was complex, with shifts to wetter or drier climates recorded in different regions (Zachos et al., 2008). Mid-latitude environments that were warm and wet were marked by a decrease in precipitation that preceded or occurred during the PETM (Garel et al., 2013; Kraus et al., 2013; Wing et al., 2005). However, proxy evidence from wet-temperate high-latitude environments such as the Canadian Arctic, Spitzbergen and North Sea, reveal positive increases to both temperature and precipitation (Eldrett et al., 2014; Greenwood et al., 2010; Uhl et al., 2007; West et al., 2015, 2020).

The seasonality of precipitation, or the availability of water, remain important aspects of paleohydrological research, particularly during greenhouse climates. Recently, global climate models have reconstructed precipitation patterns for the early Eocene and identified the existence of robust seasonal precipitation patterns within much of the middle and lower latitudes (e.g., Huber & Goldner, 2012), whereas the high latitudes were instead modeled as having a low precipitation seasonality, or considered “ever-wet” (Carmichael et al., 2016; Huber & Goldner, 2012). Leaf physiognomic analyses from Stenkul Fiord (Ellesmere Island) also showed that estimates of mean annual precipitation and growing season precipitation indicated a more even distribution of annual precipitation (West et al., 2015). In contrast, high-resolution carbon isotope analyses of fossil wood from Stenkul Fiord showed a summer peak in precipitation and characterized the region as “monsoonal” (Schubert et al., 2012).

## 2.2. Geologic Setting

Ellesmere and Axel Heiberg islands are located in the Canadian Arctic Archipelago within the Nunavut territory of Canada (Figure 1). These islands are at present positioned from 77°N to 82°N, and were approximately 2°



**Figure 2.** Simplified stratigraphic column of relevant Eureka Sound Group formations with sample locations. Approximate sampling intervals indicated by colored bars (blue) for each locality, and include: Stenkul Fiord (US435, US436, US438), Split Lake (US444), Axel Heiberg (Strand Fiord [SD], US176; Buchanan Lake [BL], US119), Strathcona Fiord (US422), Fosheim Peninsula (Hot Weather Creek, US107, US108; Mosquito Creek, US196; Fosheim Anticline, US111, US191, US251), and Lake Hazen (US261). Pink shaded regions indicate major climatic events (e.g., Westerhold et al., 2020), base column and depositional environments adapted from West et al. (2019) and Miall (1986).

further south ( $75^{\circ}\text{N}$  to  $80^{\circ}\text{N}$ ) from 66 to 40 Ma (Irving & Wynne, 1991; Jahren, 2007; West et al., 2019). At these latitudes, the islands experienced continuous darkness during winter and continuous light during summer, with short spring and fall transitions. The Sverdrup Basin (approximately 1,000 km by 350 km) spans the northern Canadian Arctic Archipelago, including Axel Heiberg Island and most of Ellesmere Island, and contains up to 13,000 m of strata of Paleozoic to Cenozoic age (Figure 1; Núñez-Betelu et al., 1994). The clastic sediments that were deposited in the Sverdrup Basin during the Paleocene and Eocene comprise the Eureka Sound Group (ESG). Our examined suite of samples is from the ESG and were deposited and deformed during burial and uplift phases of the Eurekan Orogeny (Dewing & Obermajer, 2011). As a result, many of the carbonates of the ESG experienced deformation and heating, which is important context when applying the isotopic data from these ESG carbonates to climate or burial history reconstructions.

### 2.3. Eureka Sound Group

The stratigraphy of the ESG was recently reviewed by West et al. (2019), which simplifies various nomenclature conventions and offers a comprehensive overview of the refined stratigraphy. The late Paleocene to early Eocene samples analyzed here are from the Mount Moore, Margaret, and Mokka Fiord Formations. Outcrops of these formations are present in many locations across Ellesmere and Axel Heiberg islands, with ages constrained by lithostratigraphy, biostratigraphy, magnetostratigraphy, and U-Pb zircon dating (Figure 2; Eberle & Greenwood, 2012; Harrison et al., 1999; Miall, 1986; Reinhardt et al., 2013; Ricketts, 1986; Ricketts & Stephenson, 1994; Sudermann et al., 2021; Tauxe & Clark, 1987; von Gosen et al., 2019; West et al., 1977, 1981, 2019). Because units are diachronous based on the location and degree of tectonic denudation experienced, a more precise age control beyond late Paleocene to early Eocene cannot be defined and samples could have varying or overlapping ages between 59 and 48 Ma (Figure 2). Unlike the other samples, the Buchanan Lake Formation on Axel Heiberg is considered middle Eocene, with an age approximation of about 45 Ma (Ricketts & McIntyre, 1986; Ricketts & Stephenson, 1994; Eberle & Storer, 1999; Eberle & Greenwood, 2012; Figure 2). Overall, the lack of a composite stratigraphy for the ESG and these sites specifically means we are unable to place our measured samples

stratigraphically or chronologically with precision, and therefore choose to group them by larger time period (late Paleocene/early Eocene vs. middle Eocene).

On Ellesmere and Axel Heiberg islands, ESG strata from the late Paleocene to middle Eocene are dominated by fluvio-deltaic, floodplain, and paludal deposits (Miall, 1986; Ricketts, 1986; Ricketts & Stephenson, 1994). Specifically, the Mount Moore/Mount Lawson Formation consists primarily of interbedded brown sand and siltstones with some plant fossils, defined as delta front/plain facies; the Margaret Formation consists of siltstones, mudstones, and coals with abundant plant fossils, defined as distributary channel/swamp facies; the Iceberg Bay Formation consists of fine sandstone channels and calcareous brown shales with thin coal seams, defined as fluvial overbank or ephemeral fluvial facies; the Buchanan Lake Formation consists of carbonaceous mudstones and thin coals, defined as fluvial overbank or ephemeral fluvial facies; and the Mokka Fjord Formation consists of silty mudstones, carbonaceous mudstones, and coals, defined as fluvial overbank facies (Figure 2; Miall, 1986). In each case, environmental interpretations of our analyzed carbonate mudstone samples where leaf fossils were recovered suggest our samples were palustrine and deposited in wet floodplain/swamp or overbank/crevasse splay deposit facies, which may have been alternately seasonally inundated and subjected to pedogenic processes (Eberle & Greenwood, 2012; West et al., 2019).

### 3. Materials and Methods

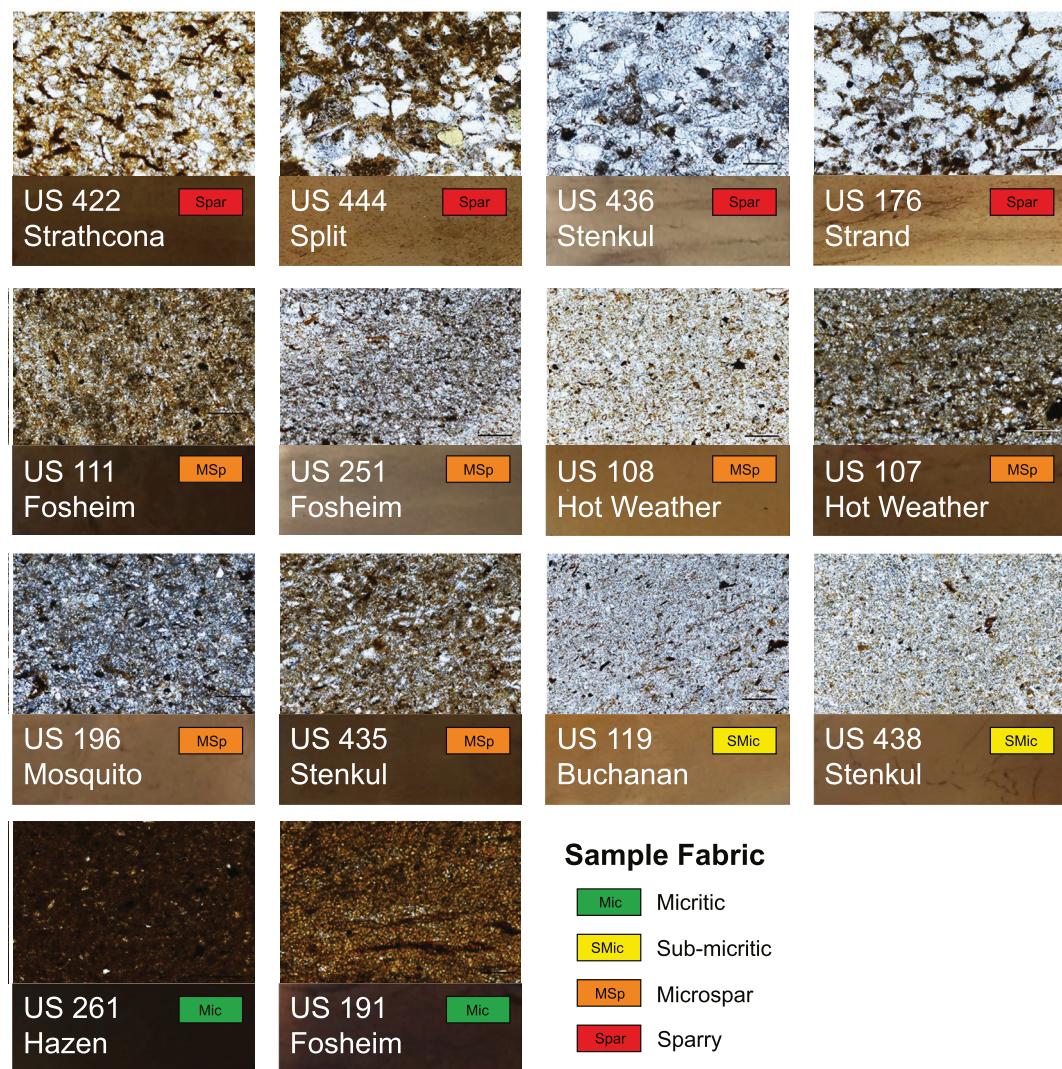
#### 3.1. Sample Collection and Evaluation

Samples were collected from paleobotanical surface quarries during multiple field seasons (1982–2004). Carbonates were associated with described fossil leaf assemblages to assign stratigraphic positions within the ESG (e.g., Eberle & Greenwood, 2012; Greenwood et al., 2010; West et al., 2015, 2019, 2020). Samples were prepared and cataloged for collections purposes within the University of Saskatchewan's Paleobotanical Collection (USPC), and a suite of 14 palustrine carbonate rocks were selected from the collection based on primary carbonate textures and geographic distribution within the ESG. Samples were selected from seven locations on Ellesmere Island (Lake Hazen, Mosquito Creek, Hot Weather Creek, Fosheim Anticline, Strathcona Fiord, Split Lake, and Stenkul Fiord) and two locations on Axel Heiberg Island (Buchanan Lake and Strand Fiord). Hand samples were cut with a rock saw to produce standard billets and sent to Wagner Petrographic for thin section preparation. Thin sections were examined under plain and polarized light on a petrographic microscope to estimate spar and micrite ratios and evaluate carbonate fabric.

To quantitatively classify samples into textural categories, a scaled percentage of spar coverage was calculated for each thin section. This approach factored in both spar coverage and crystal size, as some slides had large spar crystals and others had “micro”-spar, and provided an additional numerical value to compare between percentages of spar versus micrite and categorical designations. A random walk of 60 spar crystals was taken for each slide to approximate an average spar crystal length and width. Spar crystal dimensions were applied into an ellipse formula to calculate average spar area, which was multiplied by a general approximation of percent spar coverage for each slide. That product was then divided by a baseline of maximum crystallinity in  $\mu\text{m}^2$  (the largest average spar size at 100% coverage on the slide) to produce a scaled percentage of spar coverage. Based on the order of magnitude (0.000X, 0.00X, 0.0X, 0.X) of the scaled percentage of spar coverage (referred to herein as the Visual Alteration Coefficient [VAC]), samples were classified as micritic (Mic), sub-micritic (SMic), microspar (MSp), and sparry (Spar) respectively (Figure 3). This method quantifies visual evidence of diagenetic alteration, but it is important to note that this VAC works on the uncertain assumption that spar size and the ratio of spar-to-micrite would have an equal effect on the scaled percentage.

#### 3.2. Isotope Analyses

Carbonate clumped isotope thermometry is based on preferential bonding of heavier isotopes of carbon ( $^{13}\text{C}$ ) and oxygen ( $^{18}\text{O}$ ) together within carbonate minerals (Eiler, 2007, 2011). The heavier isotopes approach a stochastic abundance at 1000°C, while at cooler temperatures heavier isotopes preferentially “clump” (Eiler, 2007). The proportion of multiply-substituted isotopologues in a carbonate sample can be used to estimate carbonate mineral growth temperatures, and constrain paleoclimatic, paleoenvironmental, and diagenetic conditions during carbonate formation (e.g., Bristow et al., 2011; Eiler, 2011; Ferry et al., 2011; Huntington et al., 2011). Carbonates were microsampled in at least two separate locations within each billet using a carbide steel burr drill



**Figure 3.** Plain light images of thin sections showing example fabrics. Microscope image size is 700  $\mu\text{m}$  by 400  $\mu\text{m}$  in each case, and images approximate average fabric for the selected samples with no specific features highlighted. Sample numbers/locality names correspond to locations indicated in Figures 1 and 2.

bit on a Stoelting handheld micro drill, and additionally subsampled from a different location on the hand sample. Powdered samples were analyzed for stable ( $\delta^{18}\text{O}$ ,  $\delta^{13}\text{C}$ ) and clumped ( $\Delta_{47}$ ) isotopes at the Paleo<sup>3</sup> Laboratory at North Carolina State University. Samples were processed in a Nu Carb Automated Carbonate on-line preparation system at 70°C. Powdered samples (stable 500–700  $\mu\text{g}$ , clumped average 2200  $\mu\text{g}$ ) were digested with phosphoric acid (specific gravity 1.94–1.96), and resultant  $\text{CO}_2$  was cryogenically separated and passed through a Porapak Q trap held at  $-28^\circ\text{C}$ . The evolved  $\text{CO}_2$  was stored in the dual inlet alongside a  $\text{CO}_2$  working gas (compositions:  $\delta^{13}\text{C} = 4.485$ ,  $\delta^{18}\text{O} = -2.450$ ).

Sample  $\delta^{18}\text{O}$ ,  $\delta^{13}\text{C}$ , and  $\Delta_{47}$  values were measured on a Nu Perspective Isotope Ratio Mass Spectrometer configured to measure m/z ratios for masses 44–49, and are reported relative to the standard Vienna-Pee Dee Belemnite (VPDB) and the Intercarb-Carbon Dioxide Equilibrium Scale (I-CDES; Bernasconi et al., 2021). Two solid standards were run before and after each set of eight unknown carbonate samples.  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  output was referenced to IAEA standards C-1 and NBS-18;  $\Delta_{47}$  output was referenced to ETH solid standards (ETH-1, ETH-2, ETH-3, ETH-4) (Bernasconi et al., 2021). Data were processed using Easotope software (John & Bowen, 2016), using the Brand et al. (2010)  $^{17}\text{O}$  correction parameters (Daéron et al., 2016; Schauer et al., 2016) and corrected with a 70°C calcite acid fractionation of 0.066 (Petersen et al., 2019). Replicates with  $\Delta_{48}$  offset values exceeding  $\pm 2\%$  were rejected due to potential contamination by organics or hydrocarbons (Guo & Eiler, 2007; Huntington

et al., 2009). The Pierce Outlier test was used to remove statistical outliers from the stable and clumped output (Burgener et al., 2016; Huntington et al., 2009), the standard error was measured based on a 95% confidence interval, and  $T(\Delta_{47})$  were calculated using the Petersen et al. (2019) calibration.

Here, calcite  $\delta^{18}\text{O}$  ( $\delta^{18}\text{O}_{\text{calcite}}$ ) values were also converted to  $\delta^{18}\text{O}_{\text{sw}}$  using the Kim and O'Neil (1997) calcite-water isotope fractionation equation, which describes the relationship between the oxygen isotope value of a carbonate mineral ( $\delta^{18}\text{O}_{\text{calcite}}$ ), the oxygen isotope value of the source water in which the calcite formed ( $\delta^{18}\text{O}_{\text{sw}}$ ), and temperature (T) (Kim & O'Neil, 1997).

$$\delta^{18}\text{O}_{\text{calcite}} \approx \delta^{18}\text{O}_{\text{sw}} + 18.03 * (10^3 / T) + 32.42 \quad (1)$$

## 4. Results

### 4.1. Sample Descriptions

Hand samples are tan to dark brown, fine-grained, and well-indurated minor carbonate rocks (8%–45%  $\text{CaCO}_3$ ). Most hand samples are massive (no lamination to rare and very fine lamination) and have significant disseminated fine organic debris and variably abundant leaf fossils throughout. Samples lack specific pedogenic and surface alteration features, and primary micrite is mixed with recrystallized phases. In thin section, samples contain no obvious biological (e.g., shell fragments, microbial coatings) or detrital grains, nor do they exhibit unique textural features beyond simple lamination and spar growth (Figure 3). Samples appear to be exclusively calcite and have two clear generations of carbonate formation (first generation micrite and spar recrystallization) but vary in the degree of diagenetic alteration (Figure 3). Sparry samples US422, US444, US436, and US176 have occasional organics and coarse spar crystals (average spar length  $\sim 55 \mu\text{m}$ , large spar crystal length  $> 100 \mu\text{m}$ ) distributed throughout the thin section. Microspar samples US111, US107, US196, and US435 have moderate to high organic content with close to an equal ratio of microspar (average spar length  $\sim 15 \mu\text{m}$ ) to micrite and organics. Microspar samples US251 and US108 have low organic content and significant spar (average spar length  $\sim 10 \mu\text{m}$ ) distribution throughout the samples. Sub-micritic samples US119 and US438 have low organic content and fine spar (average spar length  $\sim 7 \mu\text{m}$ ) interspersed with micrite. Micritic samples US261 and US191 have very high organic content and very low spar presence (average spar length  $< 7 \mu\text{m}$ ).

### 4.2. Isotope Analyses

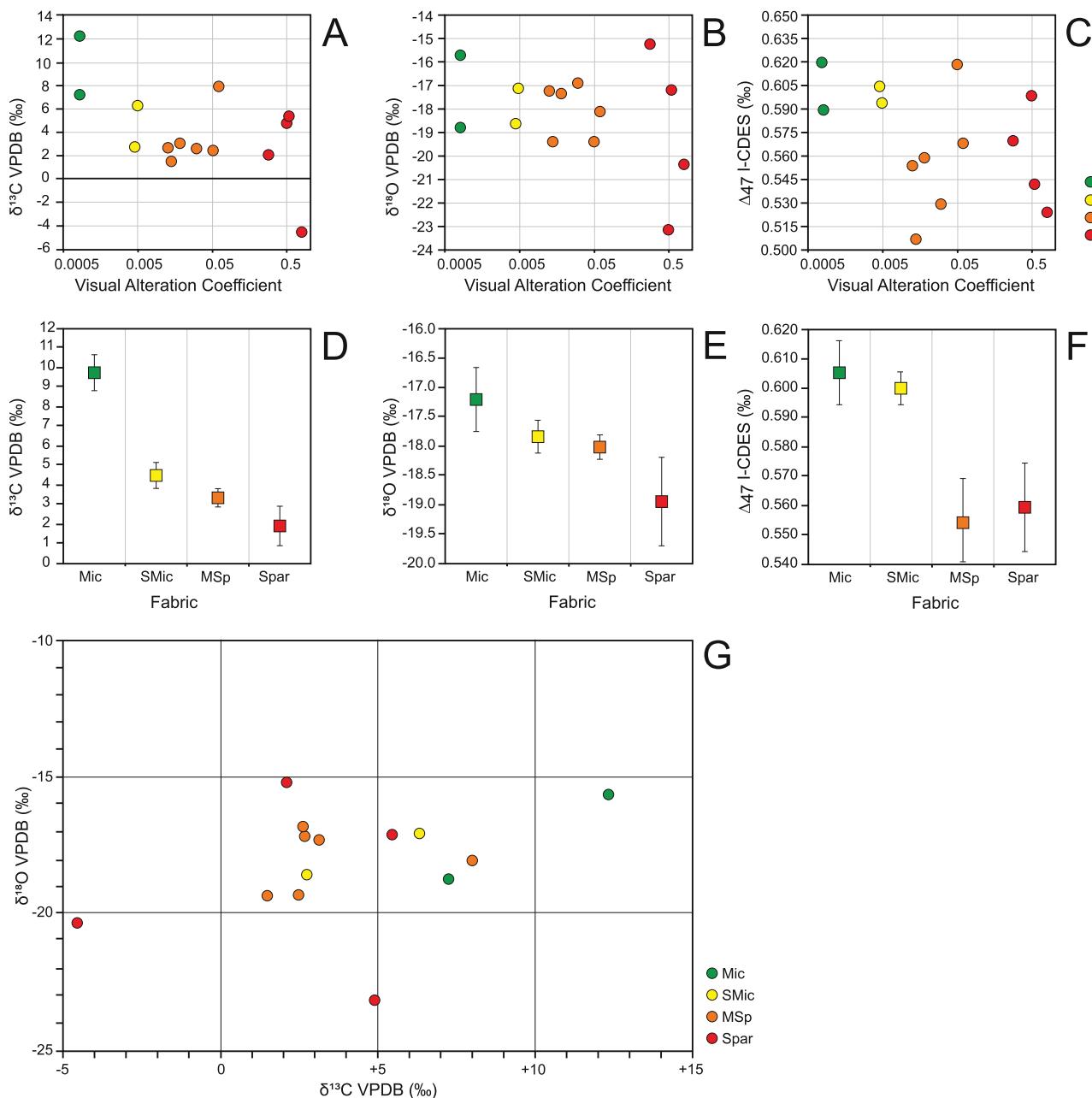
$\delta^{13}\text{C}$  isotope values range from  $-4.6$  to  $+12.3\text{\textperthousand}$  (VPDB) and  $\delta^{18}\text{O}$  isotope values range from  $-23.1$  to  $-15.2\text{\textperthousand}$  (VPDB). Samples within each fabric type (micritic, sub-micritic, microspar, and sparry) have  $\delta^{13}\text{C}$  averages of  $9.8$ ,  $4.5$ ,  $3.4$ ,  $2.0\text{\textperthousand}$ , respectively. Samples within each fabric type have  $\delta^{18}\text{O}$  averages of  $-17.2\text{\textperthousand}$ ,  $-17.8\text{\textperthousand}$ ,  $-18.0\text{\textperthousand}$ ,  $-18.9\text{\textperthousand}$ , respectively (Figure 4; Table 1).  $\Delta_{47}$  values range from  $0.505$  to  $0.621\text{\textperthousand}$  (I-CDES) with resulting temperature estimates ( $T[\Delta_{47}]$ ) from  $52$  to  $121^\circ\text{C}$ . Temperatures were on average higher for samples experiencing more significant alteration (higher VAC; Figure 4; Table 1).

## 5. Discussion

### 5.1. Sample Interpretation

Evaluation of the hand-samples and thin-sections provides evidence of the pedogenic, groundwater, and subsequent burial processes that impacted sample  $\delta^{13}\text{C}$ ,  $\delta^{18}\text{O}$ , and  $\Delta_{47}$  values. High fossil leaf preservation and lack of calcrete characteristics and surface alteration features (including desiccation features, modifications associated with roots and soil organisms, and the remobilization of carbonate and iron) serve as evidence that samples are less-developed palustrine carbonates that underwent minimal pedogenic development (Alonso-Zarza, 2003; Freytet, 1984; Marty & Meyer, 2006). Well-indurated hand samples indicate that primary micrite is mixed with recrystallized phases, and samples may have undergone varying degrees of aggrading neomorphism from pore fluid interactions (Alonso-Zarza et al., 2006).

In thin section, samples display two dominant generations of carbonate formation (first generation micrite and spar recrystallization) but vary in the degree of secondary diagenetic alteration. Consistent coarse spar distribution and equant/elongate crystal formation within microspar and sparry sample thin sections suggest they



**Figure 4.** Stable and clumped isotope results. Visual Alteration Coefficient versus carbon isotope composition (a), oxygen isotope composition (b), and clumped isotope composition (c). Isotopic averages for each fabric type with standard deviation bars (d–f). Carbon–oxygen isotope crossplot (g).

experienced burial diagenesis. The scaled percentage of spar coverage was used to evaluate the isotope data within respective fabric categories (Mic, SMic, MSp, Spar). By using a numerical classification scheme, changes in primary carbonate fabrics can be more accurately quantified to indicate how much alteration occurred due to burial and reheatting.

## 5.2. Burial and Deformation in the Sverdrup Basin

### 5.2.1. Stable Isotopes and Burial Diagenesis

In addition to the environmental processes described below (see Section 5.3), the  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values of some of our samples are influenced by diagenesis related to post-depositional burial. In order of increasing diagenetic

**Table 1**  
*Stable and Clumped Isotope Data Summary*

Sample	Location	VAC	Fabric	Replicates (n = )	Avg $\delta^{13}\text{C}$	SE ( $\delta^{13}\text{C}$ )	Avg $\delta^{18}\text{O}$	SE ( $\delta^{18}\text{O}$ )	Avg $\Delta_{47}$	SE ( $\Delta_{47}$ )	Temp (°C)	Temp error (±°C)
US119	BL	0.0045	SMic	7	2.73	0.099	-18.61	0.271	0.604	0.018	59.6	6
US444	SP	0.5	Spar	6	4.86	0.070	-23.12	0.140	0.599	0.026	62.2	9
US107	HW	0.03	MSp	7	2.60	0.097	-16.86	0.139	0.529	0.018	102.5	6
US108	HW	0.018	MSp	9	3.10	0.047	-17.31	0.035	0.558	0.033	84.2	10
US435	SK	0.06	MSp	8	7.95	0.114	-18.07	0.088	0.567	0.016	79.0	5
US436	SK	0.54	Spar	6	5.42	0.574	-17.14	0.398	0.542	0.012	94.2	4
US438	SK	0.0048	SMic	9	6.31	0.046	-17.06	0.161	0.596	0.021	63.2	7
US261	HZ	0.0008	Mic	7	12.32	0.094	-15.67	0.052	0.621	0.032	51.6	10
US191	FW	0.0008	Mic	8	7.23	0.130	-18.74	0.053	0.589	0.025	66.8	9
US111	FW	0.0125	MSp	7	2.68	0.121	-17.17	0.227	0.555	0.021	86.0	7
US251	FE	0.014	MSp	6	1.47	0.078	-19.36	0.196	0.505	0.03	120.6	10
US422	ST	0.28	Spar	8	2.09	0.024	-15.21	0.260	0.570	0.02	77.4	7
US196	MC	0.05	MSp	9	2.45	0.029	-19.33	0.187	0.618	0.023	52.9	8
US176	SD	0.8	Spar	5	-4.56	0.166	-20.32	0.803	0.525	0.001	105.6	3

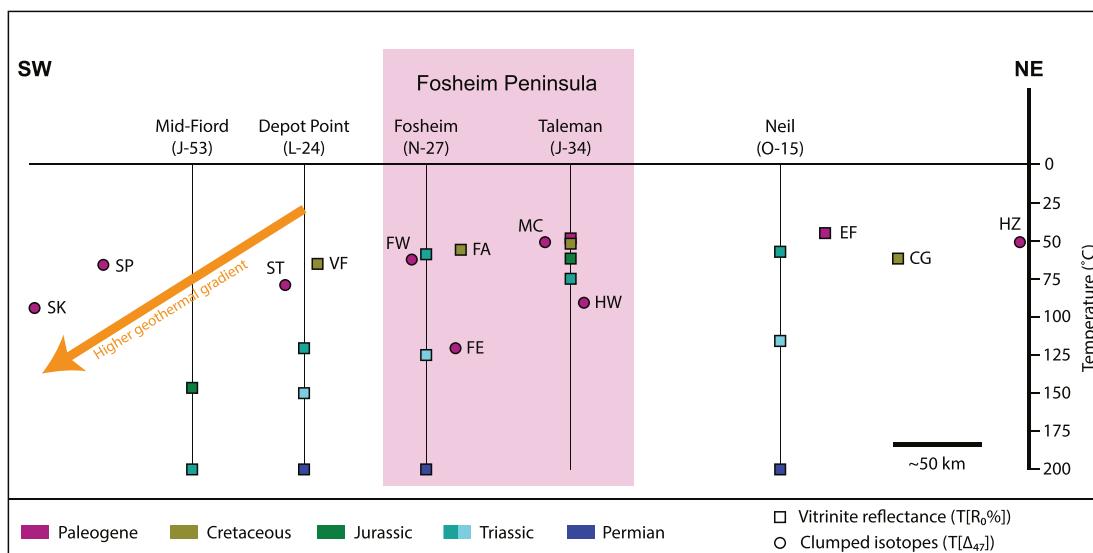
*Note.* Abbreviations for locations as in Figure 1, and fabrics as in Figure 3.  $\Delta_{47}$  values reported in I-CDES, corrected to 90°C.

alteration, the mean  $\delta^{13}\text{C}$  values for our samples decrease from 9.8‰ for Mic, 4.5‰ for SMic, 3.4‰ for MSp, and 2.0‰ for Spar, and mean  $\delta^{18}\text{O}$  values decreased from -17.2‰ for Mic, -17.8‰ for SMic, -18.0‰ for MSp, and -18.9‰ for Spar (Figure 4). Both the mean  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values display a marked overall decrease with increased alteration, showing a transition from primary micrite (original source water composition) to increasing proportions of secondary calcite spar (burial fluid composition) (Dunagan & Turner, 2004; Tandon & Andrews, 2001). Variation in  $\delta^{13}\text{C}$  within each respective alteration class likely reflects an influx of depleted carbon sourced from hydrothermal fluids equilibrated with more deeply buried carbonates; for example, deeper Permian-Triassic carbonates from these locations have  $\delta^{13}\text{C}_{\text{org}}$  of about -25 to -30‰ (Grasby & Beauchamp, 2008), and carbonates precipitated from groundwater equilibrated with these deeper rocks would have  $\delta^{13}\text{C}$  values of about -10 to -15‰ (Oehlert & Swart, 2014).

Gradual decreases in  $\delta^{18}\text{O}$  from micritic to secondary calcite samples likely reflect the temperature-dependent fractionation of  $\delta^{18}\text{O}$  as the spar crystallized out of hot diagenetic fluids with a distinct composition (Kim & O'Neil, 1997; Tandon & Andrews, 2001). Using known temperature-dependent fractionation relationships between the  $\delta^{18}\text{O}$  of calcite and source fluids (Cerling & Quade, 1993; Kim & O'Neil, 1997), we can use the methods shown above (Section 3.2) for microspar and spar samples. By substituting actual clumped isotope temperatures ( $T[\Delta_{47}]$ ) for an assumed environmental temperature (cf., Section 5.3.2), we can calculate source water ( $\delta^{18}\text{O}_{\text{sw}}$ ) values for these spar-formation fluids, which range from -2.1 to -5.2‰ (VSMOW). These values are significantly enriched in  $^{18}\text{O}$  relative to estimated environmental/meteoric waters for the region during the Paleogene (e.g., Jahren & Sternberg, 2008; Tripati et al., 2001), suggesting source fluids for spar components were either dominantly metamorphic (e.g., Hoefs, 2021) or were hydrothermal fluids undergoing significant water-rock interactions in the subsurface (see above; see also Ferry & Gerdes, 1998; Taylor & Epstein, 1962). Localized impacts of hydrothermal fluids in these sediments are unsurprising, given the regional gradient in deformation across the basin (e.g., Figure 1b; Embry & Beauchamp, 2008).

### 5.2.2. Carbonate $\Delta_{47}$ and Burial Diagenesis

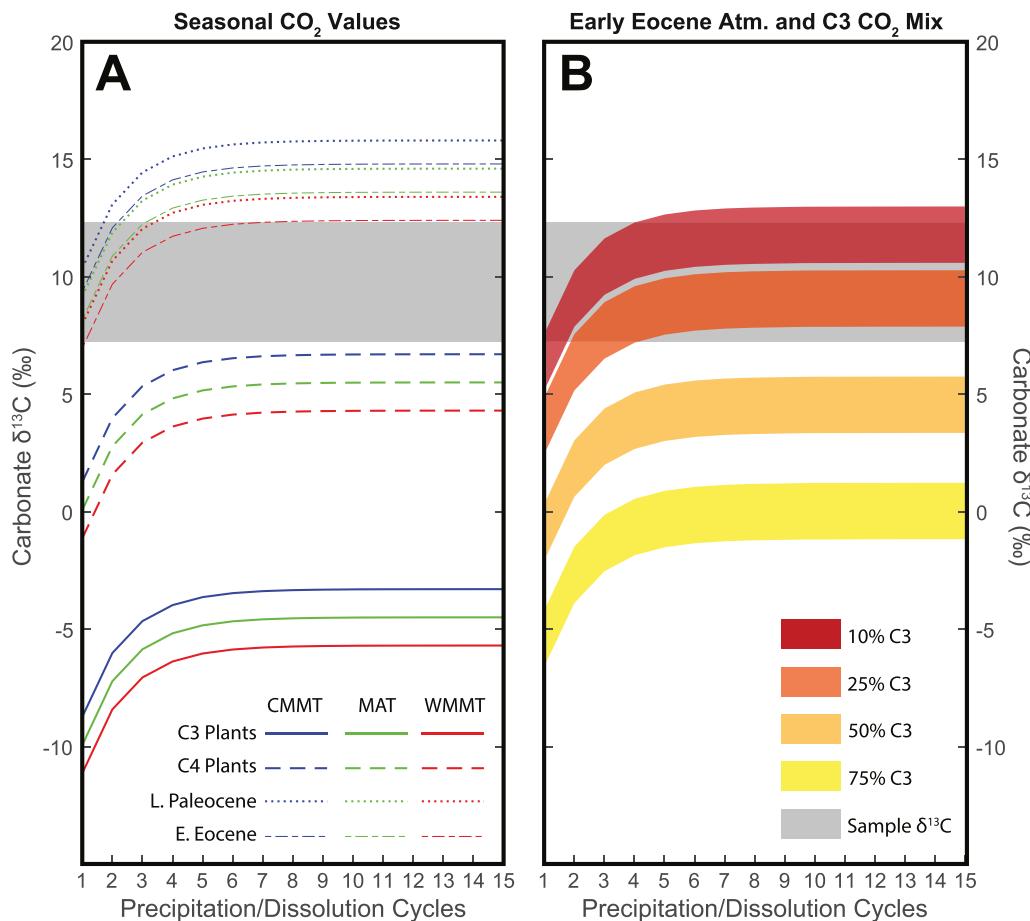
$\Delta_{47}$  ratios of 0.505–0.621‰ (I-CDES), which correspond to temperatures of 52–121°C, do not reflect Earth surface temperatures (cf., Fetrow et al., 2022). Even micritic primary samples, which did not undergo significant visible diagenetic alteration, return an average temperature of 60°C, likely due to solid-state reordering during long-term shallow burial conditions. Solid-state reordering resets clumped isotope bonds through the diffusion of C and O through the mineral lattice, changing the carbonate  $\Delta_{47}$  without affecting stable isotope values or



**Figure 5.** Spatial distribution of estimated burial temperatures across Ellesmere/Axel Heiberg Islands from vitrinite reflectance (Arne et al., 2002) and clumped isotopes (this work). Temperatures averaged for given units/ages and displayed across transect shown in Figure 1. Gray vertical lines represent well samples (names at top); other samples from outcrop (SK, Stenkul Fiord; SP, Split Lakes; ST, Strathcona Fiord; VF, Vesle Fiord; FW, Fosheim West; FE, Fosheim East; FA, Fosheim Anticline; MC, Mosquito Creek; HW, Hot Weather Creek; EF, Emma Fiord; CG, Chapman Glacier; HZ, Lake Hazen). Pink box indicates samples taken on the Fosheim Peninsula/Anticline, and orange arrow indicates region with increasing geothermal gradient (modern, assumed relict; Arne et al., 2002).

mineral texture (Henkes et al., 2014; Passey & Henkes, 2012). Our micritic sample results are consistent with low-temperature solid-state reordering, which has been shown to occur in fine-grained carbonates that experience shallow burial conditions (cf., Winkelstern & Lohmann, 2016). Winkelstern and Lohmann (2016) demonstrated that solid-state reordering of Paleogene-aged carbonates to apparent  $\Delta_{47}$  temperatures of 40–75°C occurred at relatively shallow depths (1–4 km) and was not accompanied by changes to conventional carbon and oxygen isotope values, indicating greater sensitivity of  $\Delta_{47}$  to burial diagenesis than stable isotopes (and also suggesting  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  record primary signals in micritic samples). Similarly, in our micritic samples,  $\Delta_{47}$  values were impacted by solid-state reordering (52–67°C) during shallow burial on 10<sup>7</sup>-year timescales, but conventional stable isotope values were unlikely to be affected by these conditions and show distinct compositions compared to known diagenetic carbonates (cf., Spar; Figure 4), suggesting they may be primary records. Shallow burial and low temperature heating of micritic samples is consistent with our knowledge of the depositional settings and preexisting thermal maturity data for ESG sediments at similar locations (e.g., Taleman Well; Figure 5). Vitrinite reflectance values from wells in ESG and related post-Cretaceous sediments from the Sverdrup Basin range from 0.3% to 0.6% (Arne et al., 2002), which corresponds to temperatures of roughly 50–75°C, consistent with the average (60°C) and range (52–67°C) of temperatures measured in our reordered micritic samples (Figure 5).

Samples that yielded temperatures higher than those likely experienced by ESG sediments during burial (>75°C, Figure 4; e.g., Arne et al., 2002; Jahren, 2007) were accompanied by visual evidence of diagenetic recrystallization (formation of calcite spar). Microspar and sparry samples have  $\Delta_{47}$  values indicating formation at an average temperature of 86°C (with some samples exceeding 100°C), reflecting the formation of diagenetic spar within a previously micritic texture (Figures 3 and 4). This suggests that during relatively shallow burial of ESG sediments (<2–3 km; e.g., Ricketts & Stephenson, 1994; Arne et al., 2002), heated burial fluids from deeper units likely either modified first-generation carbonates through mineralogical recrystallization, or crystallized new coarse calcite spar in pore spaces (Alonso-Zarza, 2003; Flügel, 2010), and the  $\Delta_{47}$  value (and  $\delta^{13}\text{C}/\delta^{18}\text{O}$  values; see Section 5.2.1) of the resultant calcite samples records diagenetic spar temperatures and compositions (cf., Huntington et al., 2011). Variation in spar replacement temperatures may reflect variations in fluid chemistry or sediment permeability (e.g., Huntington et al., 2011), or possible proximity to faults and deformation features (e.g., Hodson et al., 2016). Spatial variations in our measured  $\Delta_{47}$  temperatures of sparry samples appear to be related to the distribution of structural features in the region (Figure 5); this is likely related to differences in thermal conductivity and geothermal gradients across faults/subsurface structures (cf., Hodson et al., 2016), and to local variations in heat flow from groundwater and varying amounts of structural erosion (cf., Arne et al., 2002).



**Figure 6.** Carbon isotope enrichment model. (a) Evolution of carbonate  $\delta^{13}\text{C}$  through repeated precipitation/dissolution cycles under cold month mean (Cold month mean temperature), mean annual (mean annual temperature), or warm month mean (warm month mean temperature) temperature conditions (see West et al., 2020), and starting with carbon from C<sub>3</sub> plants, C<sub>4</sub> plants, late Paleocene/early Eocene (P/E) atmosphere, or middle Eocene (ME) atmosphere. (b) Modeled evolution of carbonate  $\delta^{13}\text{C}$  subject to repeated cycles of precipitation and dissolution, using a mix of late Paleocene/early Eocene (P/E) atmosphere and C<sub>3</sub> plant respired carbon. Gray shading in each panel represents range of measured carbonate  $\delta^{13}\text{C}$  in our palustrine samples.

More detailed work on spar geochemistry in these and nearby localities may allow for a more precise understanding of fluid connectivity and heat flow around structural features (e.g., Fosheim Anticline) associated with ESG deposition and more recent exhumation.

### 5.3. Stable Isotopes and Polar Paleohydrology

#### 5.3.1. Carbon Isotope Interpretations

Palustrine carbonate isotope signatures are generally intermediate between that of lake and soil carbonates; they are initially formed with compositions similar to that of a lacustrine carbonate, though they often exhibit lower  $\delta^{13}\text{C}$  values as a result of subsequent pedogenic modification (Tandon & Andrews, 2001). For lake-setting palustrine carbonates, primary  $\delta^{13}\text{C}$  values reflect the varying influences of the biogenic  $\delta^{13}\text{C}$  composition (C<sub>3</sub> or C<sub>4</sub> biomass and lake productivity) and atmospheric CO<sub>2</sub> composition (Bernasconi & McKenzie, 2007). High microbial and algal lake productivity cause primary carbonates to have  $\delta^{13}\text{C}$  values between -2 and -11‰ (Alonso-Zarza, 2003; Tandon & Andrews, 2001). Thus, the unusually positive carbon isotope values observed in our micritic samples (+7.2 to +12.3‰) and even sub-micritic samples (+2.7‰ to +6.3‰) require a mechanism that causes significant  $\delta^{13}\text{C}$  enrichment in depositional settings. The most likely enrichment mechanisms, based on the location and depositional environments, are: (a) evaporative enrichment; (b) cryogenic kinetic isotope effects; (c) anoxic methanogenesis; and (d)  $\delta^{13}\text{C}$  enrichment due to repeated dissolution-precipitation cycles.

Many pedogenic and lacustrine environments where carbonate is precipitated are described as closed or relatively evaporative, and evaporation under these conditions has been shown to develop carbon isotope enrichment via kinetic effects associated with concentration of the dissolved inorganic carbon pool (e.g., Horton et al., 2016; Ufnar et al., 2008). However, this process involves extreme enrichment of oxygen isotopes in these environments as well, resulting in strong covariance of  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  and high  $\delta^{18}\text{O}$  values (e.g., Horton et al., 2016; Talbot, 1990). Neither of these conditions are observed here (Figure 4), suggesting that evaporative enrichment alone is insufficient to explain our carbon isotope measurements.

Cryogenic kinetic isotope effects are associated with bicarbonate dehydration from a calcium carbonate solution driven by rapid freezing of the solution and simultaneous carbonate precipitation (Burgener et al., 2018). These cryogenic kinetic isotope effects result in positive  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  anomalies and negative  $\Delta_{47}$  anomalies (e.g., higher apparent carbonate formation temperatures; Burgener et al., 2018; Tripati et al., 2015). While the high  $T(\Delta_{47})$  observed in our samples may be consistent with cryogenic kinetic isotope effects, the measured  $\delta^{18}\text{O}$  values are not enriched relative to the expected environmental water  $\delta^{18}\text{O}$  composition (see below; Jahren & Sternberg, 2008). Additionally, paleobotanical and paleontological evidence shows that our sampling locations did not experience subzero temperatures for extended periods, if at all, suggesting that the rapid freezing events required to produce cryogenic kinetic isotope effects were unlikely (e.g., Basinger et al., 1994; Dawson et al., 1993; Eberle et al., 2014; Jahren & Sternberg, 2003; West et al., 2020). Based on the relatively warm paleotemperature reconstructions and the lack of positive  $\delta^{18}\text{O}$  anomalies, it seems unlikely that cryogenic kinetic isotope effects are responsible for the high primary carbonate  $\delta^{13}\text{C}$  values observed in our samples.

In uniquely hypertrophic environments, anoxic methanogenesis (i.e., where methane is being produced but not oxidized) can produce significant isotopic fractionation between organic carbon and carbonates (e.g., Conrad et al., 2009; Teranes & Bernasconi, 2005). For example, Conrad et al. (2009) found carbonate  $\delta^{13}\text{C}$  values of  $+9\text{\textperthousand}$  for their samples forming in an active methanogenic environment within the anoxic zones of a hypertrophic lake. While this range of carbon isotope values is consistent with our record, this mechanism requires intense anoxia and its observation is restricted to highly organic (near-coal) layers with heavily depleted  $\delta^{13}\text{C}_{\text{org}}$  values (e.g., Teranes & Bernasconi, 2005). Visual inspection of our samples suggests they are less carbon-rich than expected for sediments characteristic of anoxic methanogenesis (cf., Teranes & Bernasconi, 2005), and  $\delta^{13}\text{C}_{\text{org}}$  values of bulk paludal and paleosol sediments in the Buchanan Lake Formation show no evidence for methanogenic depletion (e.g., Byrne, 2005). Therefore, while some enrichment may be possible due to hypertrophic conditions by themselves ( $\sim 1\text{--}2\text{\textperthousand}$ ; e.g., Teranes & Bernasconi, 2005), it seems unlikely that this mechanism is sufficient to produce the highly enriched carbonate  $\delta^{13}\text{C}$  values observed in our sampled environments.

We suggest then that a more likely carbonate  $\delta^{13}\text{C}$  enrichment mechanism is repeated dissolution/precipitation cycles, which would increase the  $\delta^{13}\text{C}$  values without changing primary  $\delta^{18}\text{O}$  or  $\Delta_{47}$  values. Repeated cycles of carbonate dissolution and reprecipitation result in an asymptotic increase in  $\delta^{13}\text{C}$  values, with the maximum  $\delta^{13}\text{C}$  enrichment value controlled by the initial  $\delta^{13}\text{C}$  composition of the carbonate and the temperature at which the dissolution/reprecipitation occurs (Burgener et al., 2018; Clark & Lauriol, 1992; Nakai et al., 1975). We note that this process only results in net calcite  $\delta^{13}\text{C}$  enrichment if the dissolution-reprecipitation cycles occur in a closed or semi-open system with regard to the transport of  $\text{CO}_2$  between the soil water solution and the atmosphere. Salomons and Mook (1986) showed that for carbonate reprecipitation reactions where soil  $p\text{CO}_2$  is  $>0.1\text{ atm}$ , Rayleigh distillation (e.g., isotopic enrichment of the soil water solution due to  $\text{CO}_2$  degassing) dominates over isotopic exchange between soil waters and the atmosphere, and the system can be considered closed. Breecker et al. (2009) showed that even in semi-arid soils (mean annual precipitation  $\leq 550 \text{ mm yr}^{-1}$ ), soil  $p\text{CO}_2$  levels can be as high as 2.6% atm, and do not typically drop below 0.13%. This suggests that the wetter, more productive palustrine environments in our study would have had sufficiently high soil  $p\text{CO}_2$ —even during the dark winter months when soil respiration rates were lower—for carbonate dissolution/reprecipitation cycles to result in net enrichment of carbonate  $\delta^{13}\text{C}$  values.

To test whether this process can explain the  $\delta^{13}\text{C}$  enrichment observed in our samples, we modeled the  $\delta^{13}\text{C}$  evolution of carbonates subjected to repeated dissolution-reprecipitation cycles following the methods described in Burgener et al. (2018) (see also: Bottinga, 1968; Clark & Lauriol, 1992; Nakai et al., 1975). The model uses the carbon isotope fractionation factors for  $\text{CaCO}_3$  and  $\text{CO}_2$ , and  $\text{HCO}_3^-$  and  $\text{CO}_2$  (Bottinga, 1968; Mook et al., 1974), and assumed initial carbonate  $\delta^{13}\text{C}$  values based on  $\text{C}_3$  and Early Eocene atmospheric  $\text{CO}_2$  compositions to estimate the  $\delta^{13}\text{C}$  value of re-precipitated carbonate following a specified number of dissolution-precipitation events.

By accounting for enrichment, one can approximate the varying influence of plant respiration and atmospheric CO<sub>2</sub> sources on the carbon isotope values using a two-component end member mixing model (Figure 6). End member compositions were approximated as  $-25\text{\textperthousand}$  for C<sub>3</sub> vegetation, the primary vegetation type during the Eocene, and  $-5.9\text{\textperthousand}$  for late Paleocene to early Eocene and  $-6.9\text{\textperthousand}$  for the middle Eocene atmospheric CO<sub>2</sub> (Jahren & Sternberg, 2008; Schaertl & Anderson, 2005; Tipple et al., 2010; Urban et al., 2010). Cold month mean temperature was approximated at 2°C, MAT was approximated at 12°C, and warm month mean temperature (WMMT) was approximated at 22°C, based on paleobotanical methods applied to fossil leaf assemblages from these same sites (Basinger et al., 1994; Eberle et al., 2010; Greenwood & Wing, 1995; Jahren & Sternberg, 2003; Schubert et al., 2012; West et al., 2020; Wolfe, 1994). Various mixing scenarios were considered by calculating the simple weighted average between different percentages of carbonate derived from pure C<sub>3</sub> or pure atmospheric CO<sub>2</sub> (e.g., 10% C<sub>3</sub>-derived CO<sub>2</sub> and 90% atmosphere-derived CO<sub>2</sub>).

Comparing the maximum isotopic enrichment from the different temperature/CO<sub>2</sub> combinations indicates that the most significant influence on  $\delta^{13}\text{C}$  for the samples is the specific source of the carbon incorporated into the carbonates (e.g., C<sub>3</sub>/C<sub>4</sub> respiration vs. atmospheric CO<sub>2</sub>; Figure 6). Our model results suggest that carbonates incorporating pure C<sub>3</sub>-respired CO<sub>2</sub> would have final  $\delta^{13}\text{C}$  values of  $-3.3\text{\textperthousand}$  (winter precipitation/dissolution) to  $-5.7\text{\textperthousand}$  (summer precipitation/dissolution). In contrast, carbonates incorporating only early Eocene atmospheric CO<sub>2</sub> would have final  $\delta^{13}\text{C}$  values of 14.8‰ (winter) to 12.4‰ (summer). As shown in Figure 6, even after multiple dissolution/reprecipitation cycles, the  $\delta^{13}\text{C}$  value of carbonates that incorporate only plant-respired CO<sub>2</sub> would still be more negative than the observed  $\delta^{13}\text{C}$  values in our micritic samples. The observed level of  $\delta^{13}\text{C}$  enrichment in the samples suggests that atmospheric CO<sub>2</sub> was the major source of carbon for these palustrine carbonates, with a relatively minor (~10–25%) carbon input from vegetation/aquatic biomass or microbially-respired CO<sub>2</sub> (Figure 6b). Given the observed composition of fossil flora (e.g., West et al., 2020), the age of the ESG sediments, and the humid nature of the reconstructed environment, it is highly unlikely that C<sub>4</sub> or CAM plants would have been present in the region (cf., Christin & Osborne, 2014; Edwards et al., 2010); therefore, the primary source of the small amount of respiration CO<sub>2</sub> incorporated into our carbonate samples was almost certainly derived from C<sub>3</sub> vegetation (cf., Figure 6).

The results of the two-component end-member mixing model place constraints on the environmental conditions during carbonate formation. Examples of dissolution-precipitation cycles resulting in  $\delta^{13}\text{C}$  enrichment of this magnitude in modern environments are restricted to high latitude sites, such as Svalbard and the Antarctic Dry Valleys (e.g., Courty et al., 1994; Nakai et al., 1975). Modern carbonates formed in high latitude regions may experience this enrichment mechanism due to the pattern of continuous darkness during winter and shoulder-season months, resulting in low biological productivity and dry conditions (leading to carbonate precipitation with little or no plant-respiration carbon input), followed by continuous summer sunlight associated with an influx of snowmelt flooding soils and creating ponded environments (leading to carbonate dissolution; Burgener et al., 2018). Our Eocene setting would have experienced similar light conditions that would have limited plant growth and respiration when low-light conditions would have promoted plant dormancy, and as freezing conditions were unlikely the Eocene Arctic, would not have experienced significant frost or snowmelt (Eberle & Greenwood, 2012). Studies of modern pedogenic and other terrestrial carbonates have shown that seasonal precipitation variability can affect the timing of carbonate precipitation (e.g., Breecker et al., 2009; Burgener et al., 2016; Peters et al., 2013; Quade et al., 2013; Ringham et al., 2016). Specifically, Gallagher and Sheldon (2016) suggested that in regions of highly seasonal precipitation, pedogenic carbonate formation is typically restricted to the driest time of the year (regardless of the temperature of that season), particularly when the influx of water decreases long enough to allow for supersaturation of carbonate (cf., Alonso-Zarza, 2003; Alonso-Zaraza et al., 2006). Based on these observations, we suggest that our sample  $\delta^{13}\text{C}$  values and the results of our modeling experiment are most consistent with the hypothesis that during the late Paleocene to early Eocene, the Canadian Arctic hydrological regime restricted palustrine carbonate formation to the non-growing season when plant productivity was at its lowest (Figure 6), requiring a significant difference in saturation conditions between Arctic summer (growing season) and winter (period of plant respiratory dormancy) in these environments.

This suggests that the Eocene Canadian Arctic experienced some hydrological or precipitation seasonality, with more water available, or a lower evaporation-precipitation ratio, during the summer than the winter. Previous work from the Canadian Arctic established two endmember concepts for hydrological cycling during the Eocene: (a) a polar monsoon system, as indicated by summer peaks in precipitation observed in fossil wood isotopes (e.g., Schubert et al., 2012) and suggested by some model constraints (e.g., Lunt et al., 2021), and (b) an

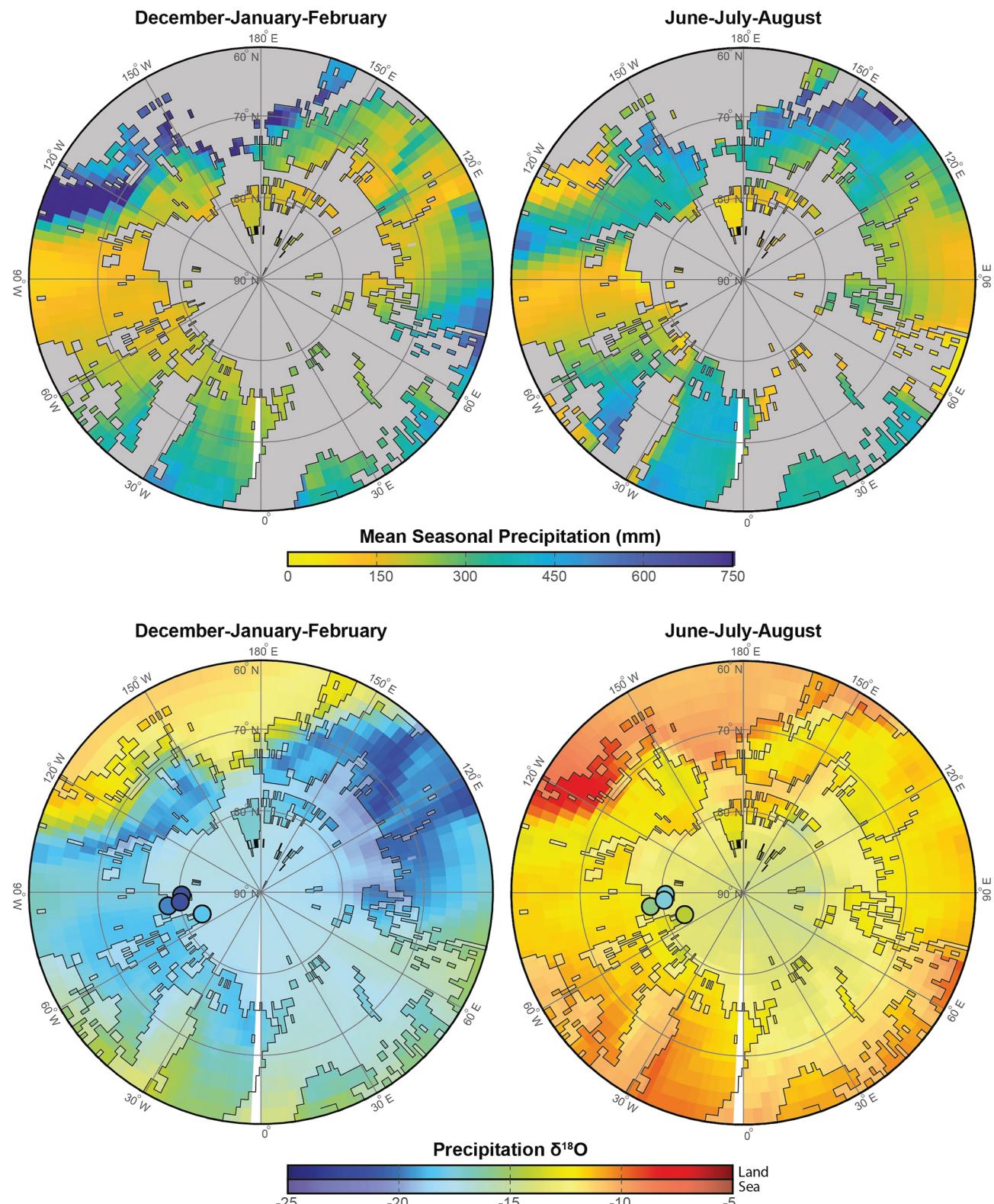
“ever-wet” equitable system, suggested as a mechanism for buffering polar warmth in other model outputs (Huber & Goldner, 2012; Kiehl & Shields, 2013) and maintaining ecosystem properties in paleontological observations (e.g., Eberle et al., 2010). However, more recent studies using climate model ensembles (e.g., Carmichael et al., 2016; Zhu et al., 2020) and comprehensive leaf physiognomy and nearest living relative analyses (e.g., West et al., 2015; West et al., 2020) have indicated that the hydrological regime of the Eocene Arctic may have been somewhat intermediate between these endmembers, where a larger proportion of precipitation is delivered in the summer months (double) and driest conditions occur during the winter/shoulder seasons (comparable to a modern/historical seasonal range; e.g., Linderholm et al., 2018; Thomas et al., 2018), without classifying as a true “monsoon” (cf., Zhang & Wang, 2008). This is supported by results from Community Earth System Model (CESM) version 1.2 as configured for the Eocene and displayed for the Arctic ( $6 \times p\text{CO}_2$  simulation; Zhu et al., 2019; Zhu et al., 2020), which shows a significantly wetter Arctic (particularly  $>70^\circ\text{N}$ ) during summer months (JJA; Figures 7a and 7b).

While the Eocene Arctic was undoubtedly wetter overall than the modern/historical regime, our data are consistent with such a seasonal pattern, with winter conditions recording carbonate deposition during a drier period of the year (when palustrine environments can be super-saturated with respect to  $\text{CaCO}_3$ ) when plants are dormant (due to temperature and/or light conditions; e.g., Basinger et al., 1994), while paired paleobotanical records from the same localities (West et al., 2015, 2020) are consistent with summer conditions recording a wetter period of the year (when carbonate dissolution occurs) when plants and soil processes are more active (due to improved temperature and light conditions). Together these records may provide a more complete picture of Eocene Arctic hydrology (cf., Carmichael et al., 2016), where summer-biased paleontological (e.g., Eberle et al., 2010) and paleobotanical (e.g., West et al., 2020) records and winter-biased isotopic records (e.g., this work) can be used together (cf., Kwiecien et al., 2022) to suggest a marked (but non-monsoonal) precipitation seasonality, which can account for seemingly disparate observations like strong isotopic signals (e.g., this work; Schubert et al., 2012) and a broad-leaved flora indicative of wet conditions (e.g., Eberle & Greenwood, 2012; Greenwood et al., 2010; West et al., 2015, 2020), as well as the presence of both palustrine coal deposits and palustrine carbonates.

### 5.3.2. Oxygen Isotope Source Water

Further evidence of seasonality can be derived from our carbonate  $\delta^{18}\text{O}$  values, where primary  $\delta^{18}\text{O}$  values in palustrine carbonates generally reflect the biogenic and temperature-dependent fractionation occurring between the calcite and original surface source water (Alonso-Zarza, 2003; Cerling & Quade, 1993; Kim & O’Neil, 1997). Evaluating the  $\delta^{18}\text{O}$  of palustrine carbonates generally provides an indirect record of local rainfall and surface water (fluvial/lacustrine) inputs, and can help filter out uncertainty about hydrological dynamics, such as mixing variations or smaller seasonal changes (Bernasconi & McKenzie, 2007). Using known temperature-dependent fractionation relationships between the  $\delta^{18}\text{O}$  of calcite and original source water (Alonso-Zarza, 2003; Cerling & Quade, 1993; Kim & O’Neil, 1997), we can estimate environmental water conditions.

For micritic and sub-micritic samples, we used an average estimate for CMMT of  $2^\circ\text{C}$  for assumed carbonate formation temperature during winter months, and an average estimate for WMMT of  $20^\circ\text{C}$  for assumed carbonate formation temperature during summer months (e.g., Basinger et al., 1994; Eberle et al., 2010; Greenwood & Wing, 1995; Jahren & Sternberg, 2003; Schubert et al., 2012; West et al., 2015, 2020; Wolfe, 1994). We use these paleobotanically-derived temperatures rather than our  $\Delta_{47}$ -derived temperatures for this calculation because the clumped isotope values have been variably reset and therefore do not represent Earth surface temperatures (see Section 5.2.2). The converted environmental water  $\delta^{18}\text{O}_{\text{sw}}$  values (VSMOW) for micritic/sub-micritic samples range from  $-14.3$  to  $-17.4\text{\textperthousand}$  for summer and  $-18.3$  to  $-21.4\text{\textperthousand}$  for winter (Figure 7). Assuming the interpretations made from the carbon isotope system are reasonable,  $\delta^{18}\text{O}_{\text{sw}}$  would not be significantly impacted by biogenic fractionation. Additionally, the dissolution/reprecipitation mechanism does not inherently affect  $\delta^{18}\text{O}$  (Burgener et al., 2018), so it is assumed that the  $\delta^{18}\text{O}_{\text{sw}}$  values are not affected by this mechanism if the source water composition has not changed significantly during the timeframe of carbonate precipitation for a given sample (years to thousands of years; cf., Alonso-Zarza, 2003). Relatively low  $\delta^{18}\text{O}_{\text{sw}}$  values are reasonable for this region and time period, because the absence of large ice sheets during the late Paleocene to middle Eocene caused global mean ocean water to have lower initial  $\delta^{18}\text{O}$  values than that during times of extensive ice cover (e.g., Zachos et al., 2008), and because  $^{18}\text{O}$  concentration decreases with increasing latitude, further reducing initial  $\delta^{18}\text{O}$  in the Arctic region (Bowen, 2010; Dansgaard, 1964).



**Figure 7.** Isotope-enabled global climate model (iCESM1.2) simulation showing early Eocene mean seasonal precipitation results (winter, DJF; summer JJA) and  $\delta^{18}\text{O}_p$  results for (winter, DJF; summer, JJA) averages ( $6 \times \text{CO}_2$ ; Zhu et al., 2020). Proxy  $\delta^{18}\text{O}_{\text{sw}}$  reconstructions from micritic samples in this study shown by colored (same scale) points.

Previous proxy estimates of environmental water  $\delta^{18}\text{O}$  composition derived from fossil wood were approximated to be around  $-15$  to  $-19\text{\textperthousand}$  (VSMOW) on Axel Heiberg Island during the middle Eocene (Jahren & Sternberg, 2008). This range overlaps with our reconstructions of  $\delta^{18}\text{O}_{\text{sw}}$  for our primary micrite samples made using either winter or summer temperatures, with theoretical winter-precipitated carbonates on the more depleted end of the range, having somewhat lower  $\delta^{18}\text{O}_{\text{sw}}$  values than those recorded during a growing season (e.g., Feng et al., 2009; Jahren & Sternberg, 2008). However, newer climate models like the CESM version 1.2 have been isotope-enabled (iCESM; Brady et al., 2019) and configured for the early Eocene (e.g., Zhu et al., 2019; Zhu et al., 2020), which allows for more detailed comparison and discrimination between summer and winter proxy reconstructions. Comparing calculated winter  $\delta^{18}\text{O}_{\text{sw}}$  with average modeled December-January-February (DJF)  $\delta^{18}\text{O}_{\text{p}}$  (Figure 7c) and calculated summer  $\delta^{18}\text{O}_{\text{sw}}$  with average modeled June-July-August (JJA)  $\delta^{18}\text{O}_{\text{p}}$  (Figure 7d), we show that our proxy results align more closely with modeled winter precipitation. While we select a  $6 \times \text{CO}_2$  simulation for this comparison based on fit described in previous work (Zhu et al., 2019), this conclusion holds for all available simulation scenarios (cf., Zhu et al., 2020).

Additionally, our relatively depleted  $\delta^{18}\text{O}_{\text{sw}}$  values agree with previous proxy (e.g., Tripati et al., 2001; Waddell & Moore, 2008) and model (e.g., Zhu et al., 2020) reconstructions of Eocene Arctic seawater conditions suggesting a closed basin with relatively fresh surface water depleted in  $\delta^{18}\text{O}$ . In order to continue to explore these important facets of the paleohydrology of high latitude regions during the Paleogene, future work should focus on well-dated and precisely mapped sections in the Arctic in order to obtain higher-resolution records of hydrological changes through time (and across the region).

## 6. Conclusions

Stable isotope results of palustrine carbonates from ESG sediments of Ellesmere and Axel Heiberg islands provide insight into hydrological conditions at high northern latitudes during the late Paleocene to middle Eocene ( $\sim 59 \text{--} 45$  Ma).  $\delta^{13}\text{C}$  isotope values range from  $-4.6$  to  $+12.3\text{\textperthousand}$  Vienna-Pee Dee Belemnite (VPDB), with carbon averages of  $9.8\text{\textperthousand}$  for micritic (Mic),  $4.5\text{\textperthousand}$  for sub-micritic (SMic),  $3.4\text{\textperthousand}$  for microspar (MSp), and  $2.0\text{\textperthousand}$  for sparry (Spar) samples.  $\delta^{18}\text{O}$  isotope values range from  $-23.1$  to  $-15.2\text{\textperthousand}$  (VPDB), with oxygen averages of  $-17.2\text{\textperthousand}$  (Mic),  $-17.8\text{\textperthousand}$  (SMic),  $-18.0\text{\textperthousand}$  (MSp),  $-18.9\text{\textperthousand}$  (Spar).  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  averages decrease with greater visual evidence of diagenetic alteration, but micritic and sub-micritic samples appear to record environmental conditions in the Eocene Arctic, which was characterized by moderate precipitation seasonality (drier winters, wetter summers) resulting in repeated dissolution/precipitation cycles for carbonate formation. Clumped isotope ( $\Delta_{47}$ ) values range from  $0.505$  to  $0.621\text{\textperthousand}$  (I-CDES), and micritic/sub-micritic samples ( $\sim 60^\circ\text{C}$ ) are consistent with low temperature solid-state reordering at expected burial depths, while microspar/sparry samples ( $> 85^\circ\text{C}$ ) are consistent with diagenetic spar formation controlled by emergence of deeper hydrothermal fluids possibly modulated by regional faulting/structures. Overall, this work highlights the utility of complex carbonate isotope data sets in distinguishing primary and post-burial signals, and contributes to a more complete reconstruction of the paleohydrology of the Eocene Arctic.

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## Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

## Data Availability Statement

All raw isotopic data is publicly available via the ClumpDB database from the Interdisciplinary Earth Data Alliance (IEDA) (Padgett et al., 2021).

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