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# Linking paleocontinents through triple oxygen isotope anomalies

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## ABSTRACT

**A central tenet of the Neoproterozoic snowball Earth hypothesis is that glaciations ended synchronously. Although this condition is borne out by U-Pb and Re-Os geochronology, the time scale of deglaciation is much less than the intrinsic error of the highest resolution dating techniques, and consequently calibrating the pace and synchronicity of biogeochemical recovery from Cryogenian glaciations remains a challenge. Given the importance of obtaining a globally synoptic view of paleoenvironmental conditions and biological evolution during these extraordinary transitions, robust correlations and chronologies are imperative. Here we extend the negative triple oxygen isotope ($\Delta^{17}O$) anomaly previously documented in ca. 635 Ma postglacial cap carbonates to two new paleocontinents, Brazil and Norway. The global footprint of this geochemical signal coupled to its short duration provides a unique time datum that can be used to cross-correlate Marinoan postglacial cap carbonate sequences and track the geochemical evolution of the oceans during deglaciation.**

## INTRODUCTION

Neoproterozoic glacial deposits are global in extent, and sedimentological and paleomagnetic data indicate that ice sheets existed at low latitudes and altitudes (Hoffman et al., 1998; Li et al., 2013). The global occurrence of sedimentologically and geochemically unique cap carbonate sequences above glacial diamictites and associated strata favor the snowball Earth hypothesis over competing explanations. This hypothesis asserts that Earth effectively froze over completely, plunging it into a highly stable climatic state dominated by the high albedo of ice. This ice albedo effect could only be overcome through the accumulation of extraordinary amounts of $CO_2$ in the atmosphere (Hoffman et al., 1998; Bao et al., 2008). Whereas the snowball Earth hypothesis was controversial, it made the key predictions that the glaciations should have been global in extent and long-lived with synchronous terminations.

Early compilations of radiometric age constraints on Neoproterozoic glaciations led some to conclude that Neoproterozoic glaciations were diachronous, and therefore inconsistent with the snowball Earth hypothesis (e.g., Allen and Etienne, 2008). However, a surge in new radiometric ages from high-precision U-Pb zircon dating and Re-Os dating of organic-rich sediments have converged to indicate that the older Cryogenian (i.e., Sturtian) glaciation initiated between 717.5 and 716.3 Ma (Macdonald et al., 2010) and terminated between 659.3 and 658.5 Ma (Rooney et al., 2014, 2015) and that the younger Cryogenian (i.e., Marinoan) glaciation initiated between 649.9 and 639 Ma (Kendall et al., 2006; Prave et al., 2016) and terminated between 636 and 634.7 Ma (Zhang et al., 2005; Condon et al., 2005; Calver et al., 2013; Rooney et al., 2015; Prave et al., 2016).

Despite new radiometric ages, most Cryogenian successions remain poorly dated. Fortunately, the geological records of the Sturtian and Marinoan glaciations and the cap carbonate sequences that were deposited in their aftermath can be distinguished via a combination of sedimentological observations, stratigraphic context, and geochemical data (Kennedy et al., 1998; Hoffman and Schrag, 2002; Halverson et al., 2005). This cap carbonate sequence begins with a transgressive systems tract (TST) that encompasses a basal cap dolostone and ends with a maximum flooding surface that commonly is within organic-rich shales and more rarely within muddy limestone. The thick overlying highstand systems tract (HST) fills the substantial accommodation space that was generated during the long-lived glaciation (Hoffman et al., 1998) but left underfilled by the unusually low sediment accumulation rates characteristic of snowball glaciations (Partin and Sadler, 2016). In contrast, the Sturtian cap carbonate sequence typically lacks a TST, beginning instead at the maximum flooding surface (Halverson et al., 2005).

Geochemical and oceanographic modeling (Crockford et al., 2016; Yang et al., 2017) imply that the post-Marinoan rise in sea level and stratification should have persisted between $10^3$ and $10^5$ yr, which is less than the current precision of radiometric dating techniques (Killingsworth et al., 2013). Given that deposition of cap dolostones is diachronous across continental margins (Hoffman et al., 2007) and that their relative timing is spatially heterogeneous due to competing factors of glacial eustasy, thermal expansion, self-gravitation, and isostatic rebound (Creveling and Mitrovica, 2014), it is difficult to generate a synoptic snapshot of the global ocean during glacial meltback and subsequent warming. However, anomalous mass-independent oxygen isotope anomalies ($\Delta^{17}O < -0.4‰$) within $SO_4$ (sulfate)-bearing minerals in multiple post-Marinoan cap dolostones, present a unique opportunity to tie geographically disparate strata together via an isochronous datum that closely approximates the Cryogenian-Ediacaran boundary. Given that the unique expression of extremely negative $\Delta^{17}O$ anomalies within post-Marinoan $SO_4$ (Bao et al., 2008, 2012; Crockford et al., 2016) is intrinsically tied to atmospheric chemistry, they must be deposited syndepositionally and the time scale of their inception and removal is likely much less than that of cap carbonate deposition. Consequently, provided the geographic footprint of this anomaly can be expanded, they can be used for unusually high precision correlation and calibration of biogeochemical evolution immediately after the Marinoan snowball Earth.

### Triple Oxygen ($\Delta^{17}O$) Isotopes

$\Delta^{17}O$ anomalies are generated by dissociating and reforming ozone ($O_3$) in the stratosphere that imparts a mass-independent enrichment of $^{17}O$ into $O_3$ and $CO_2$ and a corresponding depletion of $^{17}O$ in residual $O_2$ (Wen and Thiemens, 1993). The magnitude of $^{17}O$ depletion, denoted as a negative $\Delta^{17}O$ value [$\Delta^{17}O = \ln(\delta^{17}O + 1) - 0.5305 \times \ln(\delta^{18}O + 1)$; see the GSA Data Repository[1], is

[1]GSA Data Repository item 2018041, additional information, methods and data, is available online at http://www.geosociety.org/datarepository/2018/ or on request from editing@geosociety.org.

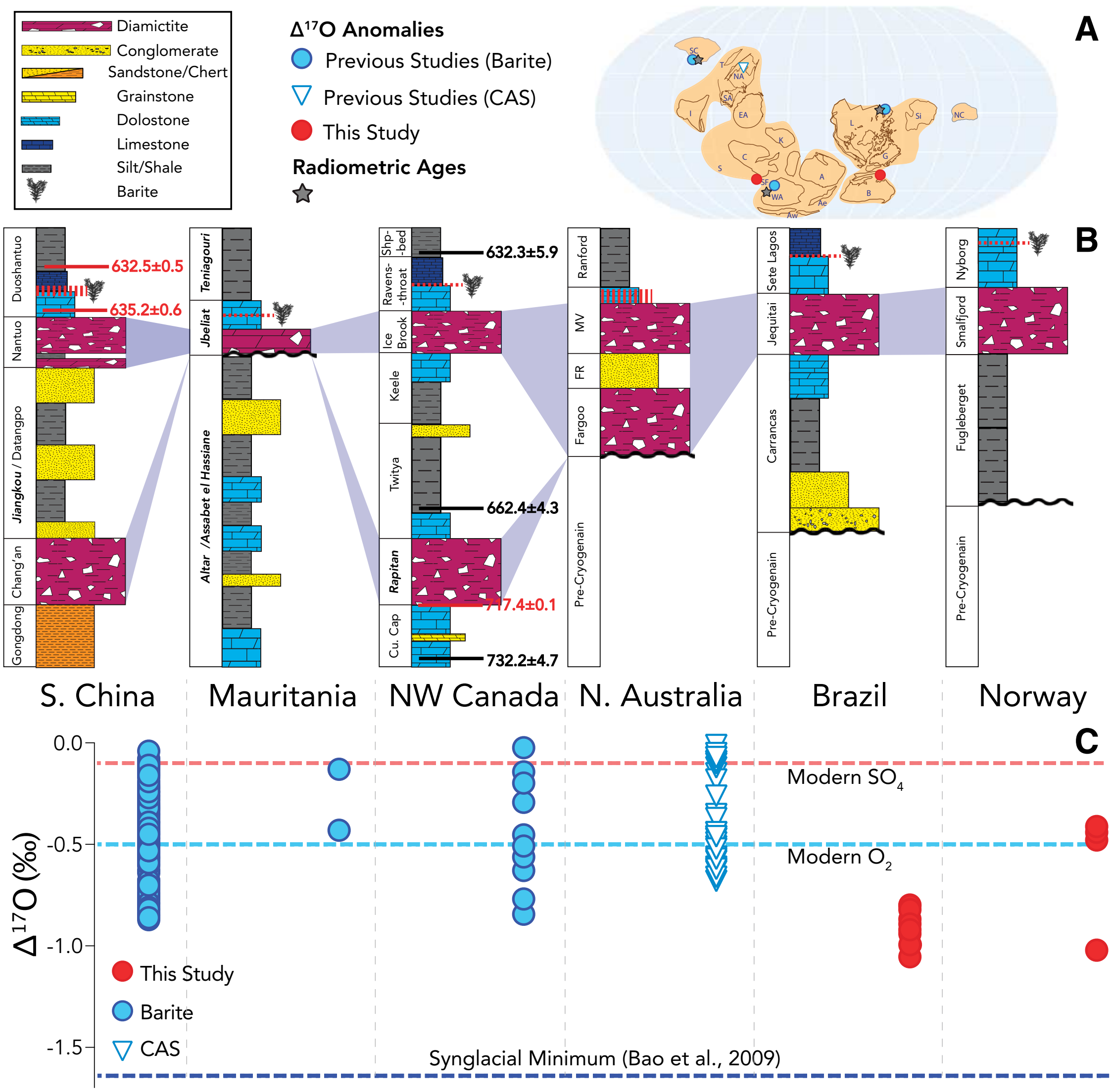


**Figure 1. Geochronological data, stratigraphy, paleogeography, and $\Delta^{17}O$ data. A: 635 Ma reconstruction map (Li et al., 2013), , sample locations (red circles), previous $\Delta^{17}O$ studies (blue circles, white triangle), and locations of radiometric ages (gray stars). B: Stratigraphic columns from (left to right) south China, Mauritania, northwest Canada, northern Australia, Brazil, and Norway, including geochronological data (U-Pb, red; Condon et al., 2005; Zhang et al., 2005; Macdonald et al., 2010; Re-Os, black; Rooney et al., 2014, 2015). Red dashed lines indicate anomalous $\Delta^{17}O$ values, and locations with barite include symbols. C: All measured $\Delta^{17}O$ data from this study and from previous works (Bao et al., 2008, 2012; Peng et al., 2011; Killingsworth et al., 2013; Crockford et al., 2016) with published barite, barite from this study, and published carbonate-associated sulfate (CAS). Modern seawater $\Delta^{17}O_{SO_4}$ is plotted as a red dashed line, modern $\Delta^{17}O_{O_2}$ as a blue dashed line, and synglacial minimum $\Delta^{17}O$ values are in dark blue. Uncertainty on all $\Delta^{17}O$ data for the total analytical procedures summarized in SI units is ±0.05‰.**

proportional to both $pCO_2$ levels and the rate of dilution via $O_2$ export from gross primary production (Luz et al., 1999). One pathway that translates atmospheric $\Delta^{17}O$ signatures to the surface environment is sulfide oxidation, where a portion of the anomaly (~8%–30%) is incorporated into product $SO_4$ (Kohl and Bao, 2011; Balci et al., 2007). Although all known post-sulfide oxidation processes will erase anomalous $\Delta^{17}O$ values and push signatures toward seawater, anomalous $SO_4$ can be preserved in the geological record (e.g., barite, gypsum, carbonate associated $SO_4$ [CAS]) provided deposition occurs before isotopic signatures are reset by microbial cycling and/or dilution by a standing $SO_4$ reservoir.

## Extending the $\Delta^{17}O$ Horizon

Of the 48 reported Marinoan glacial and immediately postglacial sequences, five bear anomalous $\Delta^{17}O$ signatures. These anomalies occur in syn-Marinoan CAS extracted from lacustrine carbonates in the Wilsonbreen Formation in Svalbard (Bao et al., 2009), post-Marinoan CAS in the Moonlight Valley cap dolostone of northern Australia (Bao et al., 2012), and most commonly in enigmatic barite horizons deposited at or near the top of cap dolostones from the Jbeliat Group of Mauritania, the Doushantuo Formation of south China, and the Ravensthroat Formation of northwestern Canada (Fig. 1; Bao et al., 2008; Crockford et al., 2016). Barite-bearing horizons typically occur discontinuously on paleotopographic highs at the transition from cap dolostones to deeper water shale or carbonate facies and range from a few millimeters to tens of centimeters in thickness as either seafloor cements or diagenetic crusts (Hoffman et al., 2011). While genetic models remain debated (Shields et al., 2007; Zhou et al., 2010), a likely scenario for barite precipitation is through mixing of segregated pools of Ba and $SO_4$ that arose as a result of postglacial density stratification. This explanation is consistent with the delayed appearance of $\Delta^{17}O$ anomalies in post-Marinoan strata while isotopically anomalous $SO_4$ accumulated during the deposition of the TST (Crockford et al., 2016).

We measured $\Delta^{17}O$ values from post-Marinoan seafloor barites in two new localities, the Sete Lagoas Formation (lower Bambuí Group) of east-central Brazil (cf. Caxito et al., 2012), and the Nyborg Formation (Vestertana Group) of Norway (Rice et al., 2011). Brazilian samples are typically 1–8 cm thick, display a bladed crystal habit, and occur along paleohighs on granitic basement. Norwegian barites also outcrop along basement highs and are typically bladed crystals and rosettes that form barite beds 1–30 cm thick. Although the ages of both units had previously been controversial, most recent studies suggest they are Marinoan, based on a combination of sequence stratigraphic, sedimentological, and isotopic characteristics (Caxito et al., 2012; Halverson et al., 2005). $\Delta^{17}O$ values are as low as −1.05‰ and −1.02‰ in the Brazil and Norway samples, respectively (Fig. 1). These values are intermediate between minimum $\Delta^{17}O$ values observed in synglacial carbonates in Svalbard (−1.64‰; Bao et al., 2009) and cap dolostones in south China (−0.87‰; Peng et al., 2011) and northwestern Canada (−0.84‰; Crockford et al., 2016; Fig. 1) and expand the occurrence of Marinoan $\Delta^{17}O$ anomalies to seven paleocontinents (Fig. 1).

### Neoproterozoic $\Delta^{17}O$ Anomalies Are Unique to the Marinoan Glaciation

At present, $\Delta^{17}O$ values <−0.4‰ are known only from Marinoan-aged glacial deposits or the TSTs at the base of the associated cap carbonate sequences. The interpretation of this geochemical signal has been controversial because varying gross primary production or $pCO_2$ levels can lead to the generation of anomalous $\Delta^{17}O$ values under very different atmospheric conditions (see the Data Repository). Evidence of relatively high levels of primary production in the Marinoan aftermath (Kunzmann et al., 2013) coupled to more in-depth modeling of the generation of $\Delta^{17}O$ anomalies over Cryogenian glaciations (Cao and Bao, 2013) support initial interpretations of extremely elevated $pCO_2$ levels (Hoffman et al., 1998; Bao et al., 2008; see the Data Repository). Explaining the restriction of these anomalies across Cryogenian strata to only the Marinoan glaciation, however, remains a challenge.

To date, no anomalous $\Delta^{17}O$ signals have been reported for Sturtian or immediately post-Sturtian sulfate (Bao et al., 2008, 2016), despite predicted high $pCO_2$ levels due to its longevity (~58 m.y.; Macdonald et al., 2010; Rooney et al., 2014, 2015). One explanation for this missing signal is that the post-Sturtian cap carbonate failed to capture the $\Delta^{17}O$ anomaly because its deposition was delayed until after the postglacial transgression, at which point the anomaly had disappeared. Consequently, the prediction is that unless Sturtian terrestrial $SO_4$ or a rare post-Sturtian TST that contains sulfate is discovered, no post-Sturtian $\Delta^{17}O$ anomaly should be preserved. Another possibility is that differences in duration (59 versus 5–15 m.y.) and ocean chemistry between Cryogenian glaciations may have led to different $SO_4$-Ba dynamics in the glacial aftermath. For example, hydrothermal and other sinks for $SO_4$ (Hurtgen et al., 2002) may have sufficiently drawn down marine $SO_4$ concentrations over 59 m.y. of glaciation that barite saturation was not achieved in the Sturtian glacial aftermath during the critical interval to capture atmospherically derived $\Delta^{17}O$ anomalies.

### Marinoan $\Delta^{17}O$ Anomalies Are Short Lived

The extraordinary atmospheric $pCO_2$ levels required to escape a snowball climate state (>250 preanthropogenic levels, PAL; 1 PAL = 280 ppm $CO_2$; Cao and Bao, 2013), combined with the positive ice-albedo feedback, would drive very rapid melting and prevent a protracted history of ice advance and retreat during deglaciation. $\Delta^{17}O$ anomalies are a consequence of this extreme atmospheric state, and its temporal expression is intrinsically linked to the time scale of $CO_2$ drawdown and translation from $O_2$ to $SO_4$. Hypotheses for the source of $SO_4$ captured within barite units require that either basin margins were strongly influenced by local continental weathering, which allows them to capture the isotopic signal of evolving atmospheric conditions, or a wholesale change to the isotopic composition of the global marine $SO_4$ reservoir. The similar isotopic expression and evolution in seven locations supports a globally connected reservoir carrying this signal (Crockford et al., 2016; Fig. 2).

A direct prediction of a globally connected reservoir is that sulfur isotopic signatures should correlate with $\Delta^{17}O$ values. Coupled data from the same samples display this relationship, with the most negative $\Delta^{17}O$ values corresponding to $\delta^{34}S$ values of ~20‰ and progressively less anomalous $\Delta^{17}O$ corresponding to increasingly higher $\delta^{34}S$ values (Fig. 2). This trend must reflect either dilution by a highly $^{34}S$ enriched sulfate reservoir or modification to the sulfate reservoir by ongoing dissimilatory sulfate reduction coupled to a high degree of pyrite burial. Using a wide range of plausible input and output fluxes, Crockford et al. (2016) calculated that for either case where $\Delta^{17}O$ anomalies are imparted and subsequently removed from a global marine $SO_4$ reservoir, the time scale for this anomaly must have been between $10^3$ and $10^6$ yrs. This framework allows for the range of observed values to be linked in time through the removal of the $\Delta^{17}O$ anomaly and evolving $\delta^{34}S$ signatures (Fig. 2). Importantly, if the deglacial global marine sulfate reservoir existed within a freshwater layer (Shields, 2005; Yang et al., 2017) and not a well-mixed ocean, the time frame is at the lower end of this range, i.e., ~$10^3$ yr. This conclusion is consistent with the fact that the anomaly is captured in only a small fraction of the TST, which is estimated to have lasted <$10^5$ yr, the time required to mix the strongly stratified postglacial ocean (Yang et al., 2017). Therefore, we argue that the $\Delta^{17}O$ anomaly is the most precise geochemical datum to cross-correlate basal Ediacaran strata, and offers the potential to further integrate and calibrate global geochemical signals during the deglaciation. In this regard, they are analogous to the iridium anomaly marking the Cretaceous-Paleogene boundary and similarly implicate an extreme event in Earth's history.

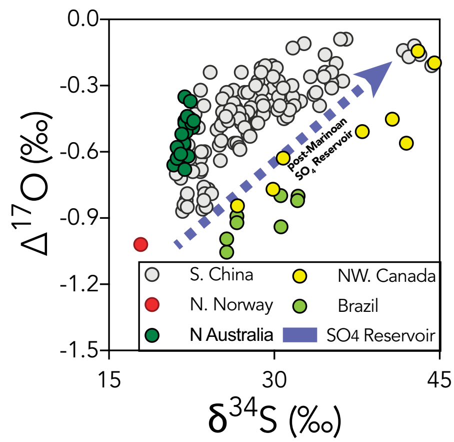


**Figure 2. Cross plot of new (this study) and previously published $\Delta^{17}O$ (*y* axis) and $\delta^{34}S$ (*x* axis) values of post-Marinoan sulfate from barites and carbonate-associated sulfate (Bao et al., 2008, 2012; Peng et al., 2011; Crockford et al., 2016). The blue line represents a proposed isotopic evolution of the global (global ocean or connected freshwater layer) $SO_4$ reservoir from initial deposition of barites to their termination. $\Delta^{17}O$ values for this study are calculated using a θ of 0.5305; however, previously published results are plotted using θ = 0.52. Errors on individual sulfur measurements are less than the plotted data points.**

## CONCLUSIONS

Correlatable datums across widespread geographic locations are paramount in reconstructing accurate temporal geochemical records to track the recovery from snowball Earth glaciations. We present new $\Delta^{17}O$ data from the Nyborg Formation of Norway and the Bambuí Group of Brazil, extending the record of Marinoan anomalies to seven paleocontinents. These new localities create a wide geographic footprint of $\Delta^{17}O$ signals that are correlatable to radiometrically dated units. The $\Delta^{17}O$ anomalies are likely unique to Marinoan-aged strata and of shorter duration than uncertainty on existing radiometric techniques. These factors make $\Delta^{17}O$ anomalies a valuable tie point for cross-correlating cap carbonate sequences from different paleocontinents and comparing other geochemical signals within them that track the rapid evolution of the Earth surface environment spanning the Cryogenian-Ediacaran boundary.


### ACKNOWLEDGMENTS

We thank Huiming Bao for providing laboratory facilities for analysis and Adam Maloof and an anonymous reviewer for providing insightful comments that improved this manuscript. Crockford acknowledges an NSERC PGS-D (Natural Sciences and Engineering Research Council Postgraduate Scholarship-Doctoral) grant and funding under Boswell A. Wing (NSERC/FRQNT—Fonds de Recherche du Québec–Nature et Technologies) and Caxito and Uhlein from FAPEMIG (Fundação de Amparo à Pesquisa do Estado de Minas Gerais, Brazil) grants APQ-00914–14, APQ-01711–14, CNPq 447449/2014–1, and PPM-00539–15.


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Manuscript received 30 June 2017
Revised manuscript received 19 November 2017
Manuscript accepted 20 November 2017

Printed in USA