Gradients in Seasonality and Seawater Oxygen Isotopic Composition along the Early Permian Gondwanan Coast, SE Australia

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Final Revision for Earth and Planetary Science Letters (Ms. Ref. No.: EPSL-D-14-01335)

Abstract

Oxygen isotope compositions of marine carbonates are commonly employed for understanding ancient temperatures, but this approach is complicated in the very distant past due to uncertainties about the effects of diagenesis and the isotopic composition of seawater, both locally and globally. Microsampled accretionary calcite from two species of the fossil bivalve *Eurydesma* Sowerby and Morris 1845 collected from sediments of Cisuralian age in high latitude marine sediments along the SE coast of Australia record cyclic seasonal fluctuations in shell $\delta^{18}\text{O}$ values during growth, demonstrating the primary nature of the isotope signal and thus allowing investigation of early Permian seawater isotopic composition and water

temperature in the high southern latitudes. The mean and seasonal range of $\delta^{18}\text{O}_{\text{carb}}$ decrease poleward across about 10° of paleolatitude ($\sim 67^\circ\text{S}$ - 77°S). The presence of co-occurring dropstones and stratigraphically associated glendonites constrain winter temperatures across the region to near-freezing, thus permitting calculation of realistic estimates of water composition and summer temperatures. Summer $\delta^{18}\text{O}_{\text{carb}}$ values indicate water temperatures between 5°C and 12°C , with warmer values at lower latitudes. The decrease in both mean sea surface temperature and seasonal amplitude with increasing latitude on the Gondwanan coast is much like that observed along high-latitude coastlines today. Calculated $\delta^{18}\text{O}_{\text{water}}$ decreases toward the pole, likely associated with an increasing contribution of isotopically light fresh water derived from summer snow-melt. The gradient in $\delta^{18}\text{O}_{\text{water}}$ is similar to that documented over a similar span of latitude on the modern SE Greenland coast. We infer the presence of a north-flowing coastal current of cold, O^{18} -depleted water that entrains progressively greater amounts of more typical seawater as it moves away from the pole. $\delta^{18}\text{O}$ values in SE Australia, however, are about 3‰ lower than those off Greenland, suggesting comparatively lower salinity water or more O^{18} -depleted glacial ice/runoff in the Permian Gondwanan high latitudes, perhaps augmented by more depleted (negative) global average seawater. Conditions in southeastern Australia during the largest of the Permian deglaciations were warmer than present-day Antarctica at similar latitudes, but may approximate those of early-mid Miocene Antarctica, with frozen winters but summers closer to 10°C .

Keywords

oxygen isotope, Permian, Gondwana, seasonality, seawater, bivalve, *Eurydesma*

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55 **1. Introduction**

56 Global climate began the transition from an icehouse to a greenhouse state during the
57 early Permian (Cisuralian, 299-272 Ma). Because continental ice sheets had contracted
58 significantly in size, the early Permian offers similarities to the waning of modern ice sheets,
59 and hence there is great interest in constraining paleoclimate over that transition (e.g., 2013).
60 Detailed stratigraphic, geochemical, and paleobotanical studies have done much to clarify the
61 conditions associated with deglaciation, but quantitative estimates of paleotemperature
62 through this interval have been fraught with the uncertainties that accompany studies of the
63 very distant past (e.g., Frank et al., 2008a; Korte et al., 2005, 2008; Rees et al., 2002). While
64 stable oxygen isotope compositions have proved invaluable for reconstructing Cenozoic
65 climate, applying this proxy in deep time is difficult due to the potential for diagenetic
66 alteration of the original isotopic compositions. This concern becomes progressively worse in
67 older samples due to the greater likelihood of exchange with diagenetic fluids and thermal
68 resetting (e.g., see Grossman, 2012, and references therein). A growing number of authors
69 have nevertheless used stable oxygen isotope values, usually of brachiopod and bivalve shell
70 fragments or sections, in the later part of the Paleozoic to interpret climate from marine
71 carbonates (e.g., Angiolini et al., 2009; Brand, 2004; 2012; 2009; Frank et al., 2008a; 2012;
72 Grossman, 2010; Grossman and Mii, 1996; Grossman et al., 1993; 2008; Korte et al., 2005;
73 2008; Lowenstam, 1961; Mii and Grossman, 1994; Mii et al., 1997, 1999; 2001; Popp et al.,
74 1986; Rao and Green, 1982; Veizer et al., 1997; 1986). These studies have vastly improved our
75 understanding of conditions during this transition, yet they still face the pervasive challenge of

demonstrating the primary nature of isotopic compositions, and most lack the spatial resolution within individual specimens necessary to address questions of seasonality (but see Ivany and Runnegar, 2010; Mii et al., 2012; Seuss et al., 2012), a component of climate that is gaining attention in studies of deep earth history.

Here, we report stable isotope data from early Permian bivalves sampled sequentially at high spatial resolution within shells. This approach has been widely used in younger specimens to resolve seasonal environmental variation recorded by organisms that produce a carbonate skeleton by accretion (see reviews by Gröcke and Gillikin, 2008; Ivany, 2012; Jones and Quitmyer, 1996; Schöne and Surge, 2012). Isotope data come from shells of the bivalves *Eurydesma cordatum* Morris 1845 and *E. hobartense* Johnston 1887 from SE Australia that span roughly 1100 km of the modern outcrop belt. *Eurydesma* is a cold-water, high-latitude, calcitic bivalve characteristic of the Permian of Gondwana (Runnegar, 1970; 1979). Specimen locations range from the northern Sydney Basin (NSB) at ~67°S paleolatitude through the southern Sydney Basin (SSB) at paleolatitude ~70°S to the Tasmanian Basin (TAS) at ~77°S paleolatitude (Klootwijk, 2009; Li and Powell, 2001; Schmidt and Clark, 2000; Scotese, 1997; Stampfli and Borel, 2002), and shells are closely associated stratigraphically with sedimentologic and mineralogic indicators of freezing conditions. Seasonally resolved records of $\delta^{18}\text{O}_{\text{carb}}$ not only afford an additional method by which to evaluate the influence of diagenesis on ancient shell carbonate (through preservation of a repeated seasonal cycle throughout ontogeny), but can also provide estimates of paleoseasonality of water temperatures on the early Permian Gondwanan coast. In addition, by sampling across a range of paleolatitudes and from sites with independent evidence for cold temperatures, it is possible to place constraints on regional

differences in water composition that might relate to latitudinal gradients associated with the Permian hydrologic cycle. Such information can provide greater clarity on the nature of climate conditions during Permian deglaciation.

2. Geologic setting and depositional environment

During the early Permian, Australia was connected to Antarctica and located close to the South Pole in the southern-most portion of Pangaea (Figure 1). Sediments considered here were deposited on the continental margin atop older Paleozoic basement or concordantly underlying Carboniferous strata. Specimens of *Eurydesma* are from shallow-water, marine, sedimentary rocks of Cisuralian (Sakmarian-Artinskian) and Guadalupian (Roadian) age that include the volcanogenic sandstones and conglomerates of the “Millfield Farm member” (informal name) of the Dalwood Group, the Allandale Formation, and the Cranky Corner Sandstone, NSB (McClung, 1980a, b; Runnegar, 1979); the pebbly sandstones of the Wasp Head Formation, SSB (Runnegar, 1970, 1980a, b; Rygel et al., 2008); and the pebbly Bundella Mudstone and dropstone-rich, fossiliferous Darlington Limestone, TAS (Clarke, 1989; Fielding et al., 2006; 2010; Isbell et al., 2012) (Figure 2; see Supplementary Information for details on stratigraphy, correlation, age control, and proximity to glacial intervals). In addition, the younger early Guadalupian (Roadian) and deeper-water Wandrawandian Siltstone (Runnegar, 1980b; Thomas et al., 2007) was also sampled. Commonly associated fossils include the epifaunal bivalve *Deltopecten*, the spiriferoid brachiopods *Trigonotreta* and *Ingelarella*, and fenestellid and trepostome bryozoans; other semi-infaunal and infaunal bivalves, crinoids, gastropods, rugose and tabulate corals, and productoid brachiopods are also variably present

(Clapham and James, 2008; Clarke, 1969, 1992; McClung, 1980a; Reid, 2010; Runnegar, 1969b; Runnegar, 1979; Runnegar, 1980a). These assemblages of organisms are characteristic of open marine shelf conditions and therefore indicate fairly normal marine salinities (Clapham and James, 2008). Brachiopods in particular are today nearly all restricted to salinities above 30 ppt (Brand et al., 2003).

Figure 1. A) Paleogeographic reconstruction for 290 Ma, based on Scotese (2001), Li and Powell (2001), and Fielding et al. (2010); New South Wales and Tasmania in dark shading. Heavy arrows mark inferred ocean surface currents from Winguth et al. (2002) and Jones et al. (2006); thinner arrow with “?” is our inferred boundary current. B) Locations of sample sites in SE Australia on modern geography (1 - Allandale, 2 - Bimbadeen, 3 – Greenlees and DM Tangorin DDH1, 4 - Wasp Head, 5 - North Head at Ulladulla, 6 - Maria Island, 7 - Derwent River at Taroona). Bowen-Gunnedah-Sydney and Tasmanian Basin systems are indicated by dark shading. (1.5 column width)

AGE (Ma)	EPOCH	STAGE	N Sydney Basin	S Sydney Basin	Tasmania
272	GUADALUPIAN	ROADIAN	BRANXTON FORMATION	WANDRAWANDIAN SILTSTONE	DEEP BAY FORMATION
283	CISURALIAN	KUNGURIAN	GRETA COAL MEASURES FARLEY FORMATION RUTHERFORD	SNAPPER POINT FORMATION PEBBLEY BEACH	BERRIDALE LIMESTONE FAULKNER AND LIFFEY GROUPS
290		ARTINSKIAN	Millfield Farm Mbr ALLANDALE FORMATION Cranky Corner Sst	WASP HEAD FORMATION	BUNDELLA FORMATION DARLINGTON LIMESTONE
295		SAKMARIAN	LOCHINVAR		QUAMBY & WOODY ISLAND SILTSTONES
299		ASSELIAN		—?— TALLONG TILLITE —?—	STOCKERS & TURO TILLITES

Figure 2. Stratigraphic relationships among lower Permian units in SE Australia. Bold-faced units are those sampled in this study. Most of the samples studied are of Artinskian age according to available U-Pb ages from eastern Australia, but have traditionally been considered older (Sakmarian). One site (UNE L901, North Head, Ulladulla, NSW) is probably early Guadalupian (Roadian) in age. See Supplementary Information for additional detail on stratigraphy, correlation, and age control. (1.5 column width)

The units sampled here, as well as underlying and overlying formations in the region, all contain indicators of cold-water deposition, including tillites, dropstones, and glendonites (Bembrick and Holmes, 1976; Crowell and Frakes, 1971; Eyles et al., 1997; Fielding et al., 2006; Fielding et al., 2010; Frank et al., 2008b; Selleck et al., 2007). The Darlington Limestone in particular contains numerous layers of dropstones, some of which are very large (Fielding et al., 2008b; 2010; Isbell et al., 2012; Reid, 2010; Runnegar, 1979). Glendonites are calcite

pseudomorphs after the mineral ikaite ($\text{CaCO}_3 \cdot 6\text{H}_2\text{O}$), an authigenic hydrated calcium carbonate that forms in sediments at temperatures below $\sim 4^\circ\text{C}$ (Bischoff et al., 1993; Marland, 1975; Selleck et al., 2007; Suess et al., 1982) and hence constrains depositional temperatures to near freezing, at least during winters. Glendonites are unlikely to directly co-occur with *Eurydesma* because they mostly form in organic-rich muds (Frank et al., 2008b), whereas *Eurydesma* preferred clean, high-energy sandy to gravelly environments (Runnegar, 1979). Nevertheless, glendonites have been found in close stratigraphic proximity to *Eurydesma* in the Cygnet-Woody Island siltstones and overlying Bundella Mudstone, Tasmania (Clarke, 1992; Fielding et al., 2010), and in the Allandale Formation, Pebbly Beach Formation, Wandrawandian Siltstone, and Branxton Formation of the Sydney Basin (Woolnough, 1910)(see additional detail in Supplementary Information), providing clear indications of cold conditions.

3. *Eurydesma cordatum* and *E. hobartense*

Eurydesma was an immobile epifaunal bivalve with heavily calcified umbones that acted as an anchor to help maintain its preferred hinge-down orientation in high energy environments with low sedimentation rates (Runnegar, 1979). While its specific taxonomic affinity is uncertain, it belongs to the Subclass Pteriomorpha (Runnegar, 1979), the group that includes the oysters, marine mussels, pectens, and arcs (Bieler et al., 2014). The thick umbones of *Eurydesma* are comprised of stable, low-Mg calcite, and offer an ideal platform for micromilling, having well-defined and expanded growth lines (Figure 3). Bivalves, including pteriomorphs, are known to precipitate calcite in isotopic equilibrium with water (e.g., Chauvaud et al., 2005; Epstein et al., 1953; Kirby et al., 1998; Klein et al., 1996; Mook and Vogel,

1968; Surge et al., 2001), making paleotemperature reconstruction possible. Earlier isotopic work on *Eurydesma* has demonstrated regular cyclic variations in shell $\delta^{18}\text{O}$ and that alternating light and dark growth bands are seasonal, with the lighter-colored regions exhibiting periods of faster growth during the warmer months and darker bands representing periods of slower growth and/or growth cessation during winter (Figure 3; Ivany and Runnegar, 2010). Here we sample two closely related and co-occurring species of *Eurydesma*, *E. cordatum* and *E. hobartense*, which differ only in the degree of convexity and thickness of the umbonal regions of the shell.

Figure 3. *Eurydesma cordatum* Morris 1845 (left) after Etheridge and Dun (1910), with inset showing cross section through the umbo of specimen MIsA from Maria Island, Tasmania. Digitized image of sampling lines is superimposed on growth banding. Note the prominent annual growth bands. (1.5 column width)

4. Ages of samples studied and relation to Gondwanan glaciation

The “*Eurydesma* beds” of the Allandale Formation (McClung, 1980a; Osborne, 1949; Percival et al., 2012; Runnegar, 1979; Walkom, 1912) and the Cranky Corner Sandstone

193 (Osborne, 1949; Slee, 1968; Stevenson, 2003) of the NSB; the Darlington Limestone at Fossil
 194 Cliff at the north end of Maria Island, Tasmania (Clarke and Baillie, 1984; Fielding et al., 2010;
 195 Isbell et al., 2012; Runnegar, 1979); and the upper part of the Wasp Head Formation, SSB
 196 (Cisterna and Shi, 2014; Gostin and Herbert, 1973; Runnegar, 1969b; Rygel et al., 2008), have
 197 long been considered approximately equivalent in age (Briggs, 1998; Clarke, 1992; Dickins,
 198 1996; Runnegar, 1969a) because of the co-occurrence of the following common and widely
 199 distributed species that are confined to the middle and late Tamarian stage of Tasmania
 200 (Clarke, 1992) and to the *Ingelarella konincki* zone in the Sydney Basin (McClung, 1978):
 201 *Deltopecten illawarensis* (Morris), *Eurydesma cordatum* Morris, *Megadesmus globosus*
 202 Sowerby, *Pyramus laevis* (Sowerby), *Ingelarella konincki* (Etheridge), *Pseudosyrinx allandalensis*
 203 Armstrong, *Strophalosia subcircularis* Clarke, *Trigonotreta stokesi* Koenig. Correlation of these
 204 cold temperate bivalve and brachiopod species with the international standard stages via the
 205 warmer temperate faunas of Western Australia has always been problematical, but a long-
 206 prevailing view is that they are Asselian-Sakmarian in age (Briggs, 1998; Dickins, 1970; Fielding
 207 et al., 2008b; Isbell et al., 2012; Retallack et al., 2011; Runnegar, 1969a; Waterhouse, 2008).
 208 However, four in-sequence Sensitive High-Resolution Ion Microprobe (SHRIMP) U-Pb ages for
 209 the Cranky Corner Sandstone (284.1 ± 2.2 Ma), underlying Beckers Formation (287.1 ± 2.4 Ma),
 210 and overlying Billy Creek Formation (285.4 ± 2.2 Ma, 285.8 ± 3.0 Ma; Claoué-Long and Korsch,
 211 2003) all fall within error in the Artinskian as currently calibrated (290.1-283.5 Ma).
 212 Unfortunately, the SL13 zircon standard used for that study was subsequently shown to be
 213 sufficiently inhomogeneous in U/Pb so as to give more variable ages than other ion probe
 214 standards, and ones that tend to be younger than IDTIMS ages by ~1% (Black et al., 2003; see

also Tucker and McKerrow, 1995). Even so, the Cranky Corner Sandstone and correlative units seem unlikely to be older than Sakmarian-Artinskian based on these SHRIMP ages, which have been adjusted downwards by 1% following Black et al. (2003; Supplemental Figure S1).

Given the age constraints above, the localities sampled for *Eurydesma cordatum* (Allandale, UCLA 7385; Bimbadeen, UNE L1113A; Cranky Corner, UNE L1292; Maria Island, UQ L3048; Wasp Head, UNE L867) must be well-separated stratigraphically from the Gondwana-wide glaciation that straddled the Carboniferous-Permian boundary and persisted into the Asselian (Fielding et al., 2008a; Fielding et al., 2008c; Montañez and Poulsen, 2013). The youngest proximal records of the P1 glacial interval in the areas studied are the Truro/Stockers/Wynyard Tillites in Tasmania (Isbell et al., 2012), the breccias at the base of the Wasp Head Formation in the SSB (Rygel et al., 2008), and a poorly exposed diamictitic interval in the lower Lochinvar Formation in the NSB (Dickins and Sullivan, 1979). In each case, our sampled horizons are separated from local glacial indicators by at least two paleontological zones, possibly corresponding to most of the Sakmarian.

Fielding and colleagues (2008a,b, 2010; Frank et al. 2015) placed the deposition of these units during and just after the waning stages of P1 glaciation and interpret overlying sediments of the lower Pebble Beach Formation and equivalents as representing another widespread glacial expansion (P2) centered in New South Wales. Isbell et al. (2012), however, characterized the interval in question in Tasmania as “postglacial” and suggested that evident ice rafting was by sea ice rather than icebergs; similar views were expressed by Clarke and Farmer (1982) and Dickins (1985). Sakmarian SHRIMP U-Pb ages from volcanics (Halls Peak Volcanics, Cawood et al., 2012; Alum Rock rhyolite, Roberts et al., 1996; Brogans Rhyodacite, Roberts et al., 2006)

that underlie even older Permian faunas in the Gunnedah Basin and southern New England Orogen (Briggs, 1998; Degeling and Runnegar, 1979; McClung, 1978) provide additional evidence for the extended interval of time (up to 8 Ma, Supplementary Figure S2) between the last of the eastern Australian ice sheets and the *Eurydesma* shells sampled in this study (Supplemental Figure S1). Whether an ice sheet is still present somewhere in eastern Australia at this time is still debated, but the deposition of these units under generally cold conditions is not. These sediments are well below the greenhouse climate crises in the Sydney Basin recognized by Retallack (2013; Retallack et al., 2011) and dated to the early middle Roadian and younger by Metcalfe et al. (2014).

5. Methods

Specimens were cut through the umbo perpendicular to growth and polished to reveal growth banding. Thin sections were prepared for the examination of shell textures and thick sections were prepared from the immediately adjacent slab for micromilling. Three or more spot samples were collected from each thick section for evaluation of diagenetic alteration using minor element (Fe, Mn) concentrations; analyses were carried out using the quadrupole ICP-MS at SUNY-Oswego. Thick sections were spot sampled for initial evaluation of stable isotope values (Supplementary Information), and 8 shells were micromilled at a resolution of 6 samples per year over 5 consecutive years. Sampling was done in the central region of the umbo in cross section to minimize potential exposure to diagenetic fluids, and near the faster-growing, juvenile portion of the ontogeny so as to exploit the more expanded growth lines and hence achieve better temporal resolution. Any evident cracks or microborings were avoided,

but several samples from obviously secondary cements filling such structures at or near the exterior surface of shells were collected for comparison with shell isotope values. Stable isotope analyses ($\delta^{18}\text{O}$ and $\delta^{13}\text{C}$) were performed at the University of Kansas using a Finnigan MAT 253 isotope ratio mass spectrometer coupled to a KIEL III automated carbonate reaction device. Paleolatitudes for each site were determined as described in Supplementary Information.

6. Results

Thin sections reveal shell fabrics with prismatic elongate calcite crystals oriented perpendicular to comarginal growth banding (Figure 4). Crystals exhibit undulatory extinction and microgrowth increments similar to primary shell textures seen in other pteriomorph bivalves such as *Pinna bicolor* (Carter and Stehli, 1980). Fe and Mn concentrations from the

Figure 4. Photomicrographs of *Eurydesma* specimens from (A) Maria Island and (B) North Head, in plane-polarized light. Note elongate complex prismatic crystals oriented parallel to the direction of growth, and changes in crystal size associated with co-marginal growth increments. (1.5 column width)

central region of *Eurydesma* shell umbos averaged 66 ± 67 ppm and 11 ± 4 ppm, respectively, and many were at or below the detection limit for Fe at 5ppm. Samples plot within the range reported for 'well-preserved' Permian bivalves and brachiopods (Korte et al., 2008; Mii et al., 2012) and for modern brachiopods (Brand et al., 2003; Came et al., 2014)(Figure 5). Modern calcitic and mixed-mineralic bivalves have similar upper limits for Fe and slightly lower limits for Mn (Cravo et al., 2007; Morrison and Brand, 1986), though still in excess of all our shell data. Crack-filling and shell-surface cements and a micro-boring infill, clearly secondary and collected for comparison with the umbonal calcite where stable isotope sampling was done, exhibit Mn >300ppm and/or Fe >1,000ppm.

Figure 5. The concentrations of Fe and Mn from our *Eurydesma* samples (solid black dots) in comparison to published data from Permian bivalves and brachiopods deemed to be well-preserved (Korte et al. 2008; Mii et al. 2012). The gray field represents the region occupied by modern brachiopods from Brand et al. (2003); modern calcitic and mixed-mineralic bivalves are comparable for Fe and slightly lower in Mn (Cravo et al., 2007; Morrison and Brand 1986). Fe and Mn concentrations of 1,000 ppm and 300 ppm, respectively, are the limits beyond which alteration is suspected. (1.5 column width)

Plots of carbonate $\delta^{18}\text{O}$ values versus distance along the growth axis define regular cyclic variation in all but one of the specimens, as expected for shell accretion that records

seasonal fluctuations in temperature (Figure 6). Values of $\delta^{18}\text{O}_{\text{carb}}$ across the study area range from 1.1‰ to -3.8 ‰ (exclusive of the shell from the Bundella Formation at Taroona, see section 7.1 below), and growth bands are associated with positive $\delta^{18}\text{O}$ values and hence winter

Figure 6. $\delta^{18}\text{O}$ versus distance along the sampled portion of the growth axis for each *Eurydesma* shell. Bimbadeen data from Ivany and Runnegar (2010). Paleolatitudes provided. NSB = North Sydney Basin, SSB = South Sydney Basin, TAS = Tasmanian Basin. (2 column width)

months. Carbon isotope values show substantially less variation within specimens, a weak tendency to co-vary with oxygen isotope values along the accretionary axis, and hover near the

published values of around +5.5‰ for the early Permian open ocean (Grossman et al., 2008; Mii et al., 1997; Mii et al., 2012; Veizer et al., 1999; Zeng et al., 2012), with shell means ranging from +4.03 to +6.63‰ (Supplementary Figure S2).

Mean winter $\delta^{18}\text{O}_{\text{carb}}$ values for each shell are computed from the most positive $\delta^{18}\text{O}$ values within each annual cycle (3-6 winters per shell), mean summer values from the most negative (2-6 summers per shell), and seasonal range from the mean difference between all adjacent summer-winter pairs (Supplemental Table S2). Winter shell values decrease with increasing latitude ($r^2 = 0.60$; Figure 7A), ranging from a mean of 0.59 ± 0.38 in the NSB to -0.72 ± 0.35 in Tasmania. There is no latitudinal trend in summer values (NSB average = -1.84 ± 0.82 ‰; SSB = -2.67 ± 0.68 ‰; Tasmania = -2.47 ± 0.27 ‰), and the range in summer shell values decreases poleward (Figure 7B). The seasonal range in $\delta^{18}\text{O}$ values, therefore, generally decreases with increasing latitude, but the trend is not without exception, as summer $\delta^{18}\text{O}$ values are more variable among locations.

Figure 7. $\delta^{18}\text{O}$ values corresponding to each winter (A) and summer (B) season of each shell, plotted by paleolatitude. Winter shell carbonate is isotopically more negative at higher paleolatitudes. Summer values are more variable at lower latitudes, suggesting greater and

more spatially variable contributions of meltwater during the summers. See Supplemental Table S1 for these values.

7. Discussion

7.1 Primary signal or diagenesis?

Critical to any paleoenvironmental interpretation based on stable oxygen isotope data is the ability to establish whether $\delta^{18}\text{O}_{\text{carb}}$ values are primary and have not been altered during diagenesis. Shell microtextures are the first line of defense in an argument for unaltered materials, but dissolution and reprecipitation under some diagenetic conditions can retain original textures with a high degree of fidelity (Budd and Hiatt, 1993; Towe and Hemleben, 1976; Turekian and Armstrong, 1961). Concentrations of Fe and Mn are more typically used to investigate alteration of marine carbonates (Brand, 2004; Brand et al., 2003; Grossman, 2012; Grossman and Mii, 1996; Popp et al., 1986). Fe and Mn are often enriched during diagenesis due to the tendency for those elements to be present at higher concentrations in pore water under anoxic conditions. A range of threshold values for Fe and Mn concentrations in brachiopod and/or bivalve calcite have been proposed by numerous authors above which diagenetic alteration is to be suspected; in this study, we adopt conservative values of 300ppm Mn and 1,000ppm Fe based on observed concentrations in modern calcitic shells (Brand et al., 2003; Morrison and Brand, 1986). It should be noted that concentrations of Fe in modern shells are influenced by other factors (Cravo et al., 2007; Morrison and Brand, 1986), and moderately elevated concentrations do not necessarily indicate alteration. Beyond these, low stable isotope values themselves can also suggest alteration by meteoric waters, with oxygen being more sensitive to alteration by meteoric fluids than carbon (Knauth and Kennedy, 2009;

Lohmann, 1988). Many otherwise pristine shells with minimal Fe or Mn from the Paleozoic nevertheless yield surprisingly depleted $\delta^{18}\text{O}_{\text{carb}}$ values (Brand, 2004; Giles, 2012; Veizer et al., 1999; Veizer et al., 1997). The observed trend toward more negative $\delta^{18}\text{O}_{\text{carb}}$ values with increasing age has led some to suggest secular change in the oxygen isotopic composition of the world oceans (Veizer et al., 1997), but this is still a contentious hypothesis (Grossman, 2012; Jaffrés et al., 2007).

Eurydesma shell textures are consistent with those observed in living bivalves (Carter and Stehli, 1980)(Figure 4), and samples of umbonal calcite exhibit Mn and Fe concentrations well within the range of modern calcitic bivalves and below our thresholds for alteration (Figure 5). Samples from clearly secondary carbonate are significantly elevated in both elements in comparison to shell concentrations (Figure 5). Oxygen isotope values are clustered in the most positive portion of the range of published Permian values for both bivalves and non-luminescent brachiopods (Figure 8, Supplemental Figure S3), consistent with primary values. Clear seasonal variation associated with growth banding is present in all but one specimen (that from the Derwent River at Taroona, Tasmania), also arguing for the retention of primary compositions. It is therefore unlikely that diagenetic alteration has been significant. The specimen from Taroona was collected from a unit intruded by a nearby Jurassic dolerite sill (Supplementary Information), suggesting a potential explanation for the more negative oxygen isotope values and lack of obvious seasonal cycles in this one shell. We do not consider it further in this analysis.

Figure 8. *Eurydesma* $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values from this study (black dots) in comparison to published Permian bivalve data from Australian samples considered primary as well as those altered to varying degrees. Shaded region encloses Permian ‘well-preserved’ (Frank et al., 2015; Korte et al., 2008; Mii et al., 2012) and modern (Brand et al., 2003) brachiopod data. Our data cluster tightly at the most positive end of both axes, while diagenesis is expected to shift values in the negative direction (Lohmann, 1988). (Column width)

The weak tendency for $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ to co-vary is only apparent within individual accretionary records and disappears when all isotope data are plotted together (Supplementary Figure S3). If covariation were the result of varying degrees of meteoric diagenesis, this would not be the case (Lohmann, 1988). Interestingly, while $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ both exhibit regular, evidently seasonal variation within shells, the relationship between the two variables is not consistent among shells or localities, and in some cases the peaks in $\delta^{13}\text{C}$ values are slightly offset from those of $\delta^{18}\text{O}$. Negative covariation along the growth axis is not consistent with diagenetic alteration but is expected to result from seasonal controls on both temperature and primary production in a well-mixed water column, and spring phytoplankton production does not wait for the water to warm, potentially resulting in offset of peaks in the two variables. Positive covariation likely reflects seasonal (summer) input of O^{18} -depleted fresh water carrying dissolved inorganic carbon derived from the decomposition of terrestrial organic matter, but

could also reflect precipitation beneath a seasonally stratified water column where warm and potentially O^{18} -depleted water remains on the surface in summer but deeper waters receive C^{12} -rich organic matter from the photic zone that is remineralized at depth. Positive covariation between $\delta^{18}O$ and $\delta^{13}C$ along a growth axis that regularly corresponds to visible growth bands is not likely to result from diagenesis, though one can envision a scenario of very low water-rock ratios and partial alteration concentrated in the faster-growing, less dense portions of the shell between dark/translucent growth bands. More than likely though, given other indicators of preservation, isotopic variation in these shells reflects primary seasonal variation in some combination of temperature, production, and salinity in these nearshore deposits.

7.2 Paleotemperatures and regional seawater composition

If shell compositions are primary, they offer the potential to calculate the temperature of the early Permian ocean on the SE Australian margin, so long as a reasonable value for $\delta^{18}O_{\text{water}}$ can be obtained. Given that these early Permian bivalves were growing at a time when Gondwana may still have been in part glaciated (Fielding et al., 2008c), we begin by assuming that $\delta^{18}O_{\text{water}}$ was close to today's global average of 0‰. The resulting annual average temperatures, as calculated from the Epstein et al. (1953) equation for mollusk calcite revised by Anderson and Arthur (1983), range from 20°C for the lowest latitudes to 24°C for the highest latitudes. Not only do these temperatures suggest increasing warmth with latitude, but they are far warmer than that presumed for ikaite formation (0-4°C, De Lurio and Frakes, 1999) and stand in stark contrast with associated cold-water indicators such as diamictites, glendonites, and dropstones in these sections. It therefore seems apparent that the local

419 $\delta^{18}\text{O}_{\text{water}}$ at these sites was lower than 0‰, and that $\delta^{18}\text{O}_{\text{water}}$ values likely decreased with
420 increasing latitude, so as to yield temperatures consistent with sedimentologic indicators and
421 the expected trend toward cooler temperatures with latitude that pervades on Earth today.

422 We therefore calculate local seawater $\delta^{18}\text{O}$ using the isotopic compositions of shell
423 accreted during winter seasonal extremes, assuming that water temperatures were near-
424 freezing at those times. *Eurydesma* growth, like that of modern high-latitude bivalves, may
425 have been restricted to a consistent minimum growth temperature associated with taxon-
426 specific physiological constraints (Schöne, 2008; Weidman et al., 1994); therefore, the winter
427 temperatures recorded are likely to be about the same at each locality. Winter temperature
428 minima in the ocean do not generally fall below -2°C because this is the point at which seawater
429 of typical marine salinity (~35‰) begins to freeze. Modern Arctic bivalves studied by Schöne et
430 al. (2005) are able to grow down to temperatures of 4°C, and bivalves around Antarctica
431 precipitate carbonate at 0°C or below (Barrera et al., 1990; 1994; Sato-Okoshi and Okoshi,
432 2008). The ikaite precursor to co-occurring glendonites precipitates only at temperatures
433 below ~4°C (Bischoff et al., 1993; Marland, 1975). Together, these observations suggest that
434 temperatures recorded by *Eurydesma* during the Permian Australian winter likely were in the
435 range of 0-4°C. Solving for $\delta^{18}\text{O}_{\text{water}}$ using the range of 0 to +4°C and observed mean winter
436 shell compositions yields local water compositions ranging from a maximum of -2.3 ‰ at the
437 lowest latitudes (assuming the higher precipitation temperature of 4°C) to a minimum of -5.4‰
438 at the highest (assuming the lower precipitation temperature of 0°C; Figure 9). These values
439 are consistent with those derived from combined Mg/Ca and oxygen isotope
440 paleothermometry on carefully chosen but undifferentiated shell carbonate in these and

related units (Frank et al., 2015). Our focus specifically on winter shell carbonate minimizes the influence of variable amounts of summer runoff and allows for resolution of latitudinal patterns that could not be discerned in this earlier study. As discussed previously, water composition is likely changing seasonally through increased input of meltwater during the warmer months, but we are unable to parameterize this contribution.

Figure 9. Best estimates of local water composition (dashed diagonal lines) off SE Australia during the Permian, bracketed by winter temperatures between 0 to 4°C (horizontal lines) and mean winter shell $\delta^{18}\text{O}$ values (vertical lines). Pale gray symbols are individual winter values of shell carbonate from the North Sydney Basin (circles), South Sydney Basin (squares) and Maria Island, Tasmania (triangles). Local water composition therefore could have ranged from ~-2.3 in the north assuming temperatures of 4 °C, to ~-5.4‰ in the south assuming temperatures of 0 °C (thick dark diagonal lines). At any single temperature of precipitation, water values vary by ~1.8‰ across the region. (2 column width)

The presence of well-preserved glendonite in the section not only constrains paleotemperatures, it also offers a complementary method for estimating the composition of high-latitude seawater. Price and Nunn (2010) used the $\delta^{18}\text{O}$ value of glendonite in combination with the conversion temperature from ikaite to calcite (about 8°C; De Lurio and Frakes, 1999) to calculate the composition of Cretaceous arctic seawater, assuming pore waters only minimally differed from seawater. We applied the same approach using $\delta^{18}\text{O}$ data from the ikaite replacement phase of glendonites published by Frank et al. (2008b). The average composition of 27 samples from the Wandrawandian Siltstone of the Southern Sydney Basin is $-2.50 \pm 1.8\text{‰}$; we therefore calculate local seawater as -4.6‰ , within the range of seawater values independently determined from our *Eurydesma* calcite in this same unit.

The gradient in mean seawater $\delta^{18}\text{O}$ values along the SE Australian coast exhibits a negative slope of about 0.14 ‰ per degree latitude (Figure 10) with an $r^2 = 0.60$, assuming a constant winter temperature of 2°C (midpoint between the two scenarios presented in Figure 9). Note that values calculated from the specimen at North Head, at 70°S, fall below the regression line. This shell is younger than the others (Roadian), and removing it from the regression increases the r^2 value to .74 (as reported in Figure 10). Given the recent suggestion by Chen et al. (2013) that deglaciation began later in the Permian than previously thought, in the Kungurian, it is possible that more negative global seawater values at the time of precipitation of this shell contributed to its depletion in comparison to the trendline from other shells. The roughly 1‰ offset is comparable to the decrease in conodont apatite $\delta^{18}\text{O}$ values between the Artinskian and Roadian reported by Chen et al. (2013) in South China, though warming temperatures also likely contributed to that drop.

Figure 10. Inferred local seawater composition along the early Permian coastline of SE Australia. Values are calculated from the winter shell $\delta^{18}\text{O}$ values for each paleolatitude, assuming a temperature of 2 °C. Winter values presume the least amount of freshwater input, as continental waters are likely to be frozen, and therefore provide the best representation of regional seawater. Regression equation and r^2 omit the isotope values from the Wandrawandian Siltstone at North Head, in pale gray, because that sample is younger than the rest. (Column width)

7.3 Reasons for lower seawater $\delta^{18}\text{O}$ values in the high latitudes

In the modern ocean, high-latitude surface-water compositions are more negative than the global mean ($\sim 0\text{‰}$) due to the influence of progressively more depleted precipitation with increasing latitude (Bigg and Rohling, 2000; Bowen and Wilkinson, 2002; Craig and Gordon, 1965; LeGrande and Schmidt, 2006). All high-latitude, open-ocean values today exceed $\sim -1.0\text{‰}$ except in the semi-restricted Arctic Ocean, which receives isotopically depleted fresh water during summer melting of polar ice and snow (Ekwurzel et al., 2001). Inferred Permian local seawater values are significantly more negative, consistent with a setting that receives additional input of isotopically more negative waters. The melting Gondwanan ice sheet is a

likely source, as the precipitation that composed it was significantly distilled (see further discussion in this section). Small amounts of meltwater combined with seawater could yield depleted values without lowering salinity enough to affect the stenohaline marine shelf community.

If the overall low values of early Permian seawater in SE Gondwana are due to fresh/meltwater mixing, then the trend of increasingly negative seawater $\delta^{18}\text{O}$ values toward the pole cannot derive from local runoff alone unless discharge was higher at higher latitudes or runoff $\delta^{18}\text{O}$ was progressively more negative toward the pole. The former is unlikely, as lower latitude sites should experience more melting and/or precipitation. The latter is possible if the source of runoff is precipitation that fell at progressively higher latitudes and hence was more distilled toward the pole. Either way however, the trend in water oxygen isotopic composition is evident in the winter, when runoff should be minimal. Mixing water masses, rather than local surface runoff, is perhaps a better option to explain the low and increasingly negative seawater $\delta^{18}\text{O}$ values with latitude.

Low oxygen isotope values and a similar negative gradient are seen in coastal waters off SE Greenland today (Schmidt et al., 1999), suggesting that this setting might offer insights about SE Australia during the Permian. The Greenland trend in water oxygen isotopic composition with latitude reflects the mixing of Atlantic Ocean water with depleted Arctic Ocean water transported south in the Polar Water of the East Greenland Current (Aagaard and Coachmen, 1968; Sutherland et al., 2009). Coriolis forces acting on this western boundary current keep the coldest (-1.9°C), lowest salinity (~ 28 ppt), and isotopically most negative ($\delta^{18}\text{O} \sim -2.5\text{‰}$) waters closest to the coastline and near the surface, and wintertime temperatures are uniformly at the

freezing point above depths of 75m regardless of location (Aagaard and Coachmen, 1968). $\delta^{18}\text{O}$ values, at least during the summer (no data are available for winter), decrease to the northeast along the coast (Schmidt et al., 1999; derived largely from Sutherland et al., 2009). At these cold temperatures, the effect of salinity on density is stronger than the effect of temperature (Sigman et al., 2004; Winton, 1997), resulting in stratification such that fresher, more negative, waters remain at the surface. While Permian open-ocean surface currents are reconstructed as generally flowing southward along SE Australia (Frank et al., 2015; Winguth et al., 2002), a similar coast-hugging brackish boundary current could have delivered polar waters to near-shore settings, with progressive dilution by isotopically more positive seawater as it flowed north and entrained offshore south-flowing waters (see arrow in Figure 1A). Upwelling of cold deep water associated with inferred offshore winds (Gibbs et al., 2002) has been invoked to explain why the SE coast of Australia was colder than the rest of Gondwana as glaciation waned (Frank et al., 2015; Jones et al., 2006). While upwelling would likely break down the observed isotopic gradient along the coast, the presence of a cold polar boundary current could account for both the gradient and unusually cool conditions. Crowell (1978) postulated the presence of such a current driven by strong polar easterlies being deflected north along the SE margin of Gondwana. North-dipping cross-bed sets in the nearshore marine Nowra Sandstone of the South Sydney Basin, just upsection from the units discussed here, suggest south-to-north sediment transport along the coast (Runnegar, 1984), consistent with such a current.

Inferred $\delta^{18}\text{O}$ values of Permian seawater are ~3‰ more negative than the waters off Greenland today, despite similar amounts of compositional change with latitude and similar inferred temperatures. Given the stenohaline nature of the *Eurydesma* fauna, it is unlikely that

this offset is due to proportionally greater contributions of runoff to the Permian coastline than Greenland sees today. More likely, Gondwanan glacier ice and meltwater were more depleted in O^{18} . The modern Antarctic ice sheet has $\delta^{18}O$ values as low as -50‰ (Morgan, 1982), and Gondwana was an even larger polar land mass with the potential for highly depleted precipitation in its interior. Assuming Permian oceans were not unlike those today and marine isotope values at ~65°S were roughly -0.5‰, a mixture with a salinity of approximately 31-33ppt (roughly 5-9% fresh water) would result from the addition of enough fresh water at -50‰ to yield calculated water values of -3.0 to -5.0‰. If meltwater or runoff were less depleted in O^{18} , proportionally more would be needed to achieve the inferred values. Salinity did not drop to the point that marine paleoecology was affected, placing an albeit poorly defined cap on the amount of fresh water mixed into coastal waters. Exceeding this limit could require a call for isotopically more negative global average seawater. If this were the case, Cisuralian paleotemperatures previously calculated from observed $\delta^{18}O_{carb}$ values would be systematically too warm (e.g., see Giles, 2012).

7.4 Paleoseasonality

The seasonal range of $\delta^{18}O_{carb}$ decreases with increasing latitude. These values can be used to infer patterns in temperature seasonality on the early Permian Australian margin. Given the typical attenuation of seasonal extremes by shell growth pointed out by Williams et al. (1982) and addressed by Wilkinson and Ivany (2002) and Goodwin et al. (2003), these are likely to be conservative estimates because the full winter portion of the seasonal signal may not be represented. Because winter water temperatures are presumed to be near freezing at

568 all localities, the decrease in the seasonal range of $\delta^{18}\text{O}_{\text{carb}}$ with increasing latitude must be due
569 to a drop in summer temperatures toward the pole and/or to increased amounts of run-off at
570 the lower latitudes in the summer. Local Permian water compositions calculated from shell
571 winter $\delta^{18}\text{O}$ values combined with an estimated minimum recorded temperature of 2°C allow
572 for an approximation of summer temperatures and hence the seasonal range of temperatures
573 recorded by each specimen, under the admittedly simplifying assumption that local $\delta^{18}\text{O}_{\text{water}}$ is
574 fairly constant throughout the year. Permian shallow marine summers are generally warmer,
575 and hence seasonality is greater, at lower latitudes (Figures 6,7); North Sydney Basin summers
576 average 10.5°C while Tasmanian Basin summers average 7.7°C. Summer temperatures are also
577 more variable in the north, with some years reaching temperatures as warm as 17°C, while
578 summers span a range of only 2.2°C in Tasmania (7.1 to 9.3°C).

579 These early Permian summer water temperatures, and hence seasonal ranges, are on
580 the high end of or exceed those at similar latitudes (north or south) today (World Ocean Atlas
581 2009, Locarnini et al., 2010). More than likely, increased freshwater runoff during the summers at
582 the lower latitudes is magnifying the apparent summer temperatures and seasonal
583 temperature range inferred from carbonate $\delta^{18}\text{O}$ values. Nevertheless, summers warmer than
584 might be expected in SE Gondwana, given the presence of a waning ice sheet on the continent,
585 could result from the influence of a large land mass in the Southern Hemisphere at this time
586 (Crowley et al., 1986), so long as portions of the continent were ice-free and so had lower
587 albedo. Glossopterid-dominated cold-temperate floras in SE Gondwana (Rees et al., 2002) and
588 our observation of large fossil trees in the South Sydney Basin nearshore marine section
589 suggests this to be the case. Coastal temperatures such as those reported here would be more

subject to seasonal continental albedo effects than open ocean temperatures. The degree to which ^{18}O -depleted freshwater runoff is augmenting calculated summer temperatures is not possible to constrain at this time.

7.5 Perspectives on high-latitude settings

Our isotope-based temperature estimates are consistent with a cold, but not frigid, polar setting. Glendonites and dropstones require freezing winters and floating ice, but the presence of driftwood in the section indicates trees on nearby land and hence summers that reach or exceed $\sim 10^{\circ}\text{C}$ (Rees et al., 2002). Today's Antarctic coasts, with water temperatures hovering near zero year-round, is therefore not the best analog for the Cisuralian of Australia, despite similar latitudes. A comparison with somewhat warmer periods of Antarctica's glaciated history, such as the early-middle Miocene (Burdigalian) when summer water temperatures may have been as warm as 11°C (Feakins et al., 2012; Lavelle, 2001; Shevenell et al., 2008; Warny et al., 2009), is perhaps more appropriate. During this interval, Antarctic ice volume varied significantly, dropping to as little as 25% of the present East Antarctic ice sheet (Pekar and DeConto, 2006), and woody plants reappeared on the continent (Warny et al., 2009), not unlike that inferred for the mid-Cisuralian.

Aspects of the Australian marine fauna, too, are better interpreted in this light. Ever since Etheridge and Dun (1910) carried out the first in-depth study of *Eurydesma*, the massively thickened shells have seemed anomalous for circumpolar conditions: "The umbonal region of the valves in *Eurydesma* is of extraordinary thickness, and it is interesting to note, as Mr. Charles Hedley has pointed out, that all recent bivalves comparable in valve thickness occur in

tropical waters” (Etheridge and Dun, 1910, p. ix). In addition, several species of eastern Australian bivalves (including *Eurydesma*), gastropods, and brachiopods are relative giants for Paleozoic seas even at lower latitudes. *Eurydesma* grew to a maximum size of 15 cm, with umbones up to 6 cm thick. The unrelated but co-occurring anomalodesmatan bivalve *Myonia corrugata* achieved similar sizes and valve thicknesses (Supplemental Figure S4B; Runnegar, 1979). One specimen of the inoceramiform pteriomorph bivalve, *Aphanaia* de Koninck, is 41.5 cm in length, and 18 species of bivalves, gastropods and brachiopods were larger than 10 cm in size (Runnegar, 1984, fig. 2). Modern polar bivalves tend to be small and thin-shelled (e.g. www.arcodiv.org/seabottom/Bivalves.html). If body size is expressed as $\text{Log}_2(\text{SQRT } L * H)$, where L and H are shell lengths and heights (Krug et al., 2010), then 23 eastern Australian Permian shells (bivalves, brachiopods, gastropods) exceeded a Log_2 size of 6, compared with only a single extant species of Antarctic bivalve (Krug et al., 2010, fig. 3). Likewise, modern polar taxa are drab in color, while at least three of the Australasian Permian species, including *E. cordatum*, were brightly colored (Percival et al., 2012, fig. 7; Runnegar, 1979, fig. 1B; Waterhouse, 1963, pl. 102 fig. 4)(Supplemental Figure S4D). To the degree that temperature is responsible for the differences in size and color, these observations together further suggest that the early Permian high southern latitudes were not as cold as those today. Indeed the Miocene again presents a reasonable analog, as some Miocene Antarctic bivalves are strikingly similar to common Permian ones [e.g., Miocene *Australochlamys anderssoni* (Beu and Taviani, 2014, fig. 3D) and Permian *Deltopecten mitchelli* (Waterhouse, 1987, pl. 4, fig. 14)].

8. Conclusions

Although a number of authors have attempted to estimate Permian temperatures from oxygen isotope values of biogenic calcite, all share the difficulty of estimating water compositions in their chosen settings. We present an alternative approach that relies on independent evidence for temperature derived from the sedimentary record and on seasonally resolved isotope data preserved in accretionary shell carbonate. Inferred freezing winter minimum temperatures and measured carbonate oxygen isotope values allow determination of early Permian water composition along the SE coast of Australia, revealing a 1.3‰ decrease over ~10° of increasing paleolatitude, a trend similar to that off of the coast of Greenland today. Calculated water values are significantly more negative, reflecting the contribution of isotopically more negative runoff from the Gondwanan continent. It should be possible to use this gradient to better estimate $\delta^{18}\text{O}$ -derived temperatures from previously recovered specimens at similar paleolatitudes along SE Australia. Equatorward, there should be a point where marine wintertime carbonate $\delta^{18}\text{O}$ begins to decrease, marking the point where warming temperatures, rather than decreasing polar runoff, become the dominant influence on the gradient. Improved high-resolution sampling across a wider range of Permian paleolatitudes and geographical locations, as well as progress toward independent controls on paleotemperatures, will help to better constrain Permian Ocean chemistry and climates.

9. Acknowledgements

Dan Petrizzo is gratefully acknowledged for collaboration on the field portion of this study. Special thanks go to Peter and Kaye Makeig (Bimbadeen), Warwick Denshire (Greenlees), Steve Hall, Simone Meakin, and Ian Percival (Geological Survey of New South

Wales), Matthew Billings (Hunter 8 Alliance), Phil Smart (Mollymook), Mike Clarke (Taroona), Trevor and Pat Hanigan (Derwent Estate Wines), Michael Comfort (Primary Industries, Parks, Water and Environment, Tasmania), and Pete Lingard (Maria Island National Park) for assistance, for access to the Allandale project, the DM Tangorin 1 core, and field sites, and for permissions to sample and collect. We thank Paul Tomascak at SUNY-Oswego for help with ICP-MS work, David Linsley at Colgate University for preparing the thin sections, Jack Hietpas and Melissa Hicks for help interpreting textures, Maddie O'Connor for help in sampling, and Artem Kouchinsky for some isotopic analyses carried out at UCLA in the E.D. Young Laboratory. All other stable isotope analyses were done at the Keck Paleoenvironmental & Environmental Stable Isotope Laboratory at the University of Kansas. This work benefited from meaningful discussions with Scott Samson, Bruce Wilkinson, and Paul Tomascak, and from substantive and thoughtful comments by three anonymous reviewers. Research was supported by the Department of Earth Sciences at Syracuse University and a Stephen Jay Gould Research Grant from the Paleontological Society (to JAB), the American Federation of Mineralogical Societies and the College of Arts and Sciences at SU (to LCI), the NASA Astrobiology Institute and the Committee on Research at UCLA (to BR). Collections used in this study were assembled at the University of New England, Armidale and UCLA by BR with financial assistance from the Australian Research Council, Dolf Seilacher (Tübingen), and the U.S. National Science Foundation.

Associated Supplemental Files:

Beard et al. Supplementary Information.doc; Beard et al suppl data file.xlsx

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