Sclerochronological Analysis of Archaeological Mollusc Assemblages: Methods, Applications and Future Prospects

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Abstract

Accreting skeletal tissues found in bone, teeth, otoliths and molluscan shell act as sensitive recorders of local environmental and climatic conditions. Owing to their robust nature, ubiquity and abundance in the archaeological record as well as the potential for high-resolution data acquisition, the accreting skeletal tissues of archaeological molluscs are increasingly employed as palaeoenvironmental proxies. Researchers have chiefly utilised such proxies to extend instrumental records of environmental conditions through palaeoenvironmental reconstruction and explore the impact of environmental and climate change on human populations. However, the use of environmental proxies from the archaeological record can be hampered by a number of methodological challenges including inadequate sampling strategies, appropriate calibration, the use of inappropriate proxies and the broad extrapolation of localised results. This paper reviews the use of molluscan shell from archaeological contexts as palaeoenvironmental proxies. We focus on the application of sclerochronology – a suite of high-resolution physical and geochemical data recovery methods widely used in conjunction with molluscan shell. This paper presents an overview of the potential of these techniques in approaching more nuanced understandings of human-environment interactions and how they can be more successfully incorporated into archaeological research.

Keywords

Palaeoenvironmental proxy, human-environment interaction, palaeoenvironmental reconstruction, sclerochronology, stable isotope analysis, trace element analysis

1. Introduction

Archaeologists are increasingly citing environment and climate as important variables impacting human decision-making in prehistory (e.g. Brockwell et al. 2013; Burchell et al. 2013c; Faulkner 2013; Hallmann et al. 2013; Hiscock 1999; Nunn 2003; Rowland 1999; Sim and Wallis 2008). Gaining increased understandings of the environments with which past populations engaged is therefore an integral first step towards more accurate interpretations of human behaviour. However, as large-scale palaeoenvironmental reconstructions can be geographically removed from contexts of interest (Shulmeister 1992), archaeologists must employ proxies which are tied more directly to the archaeological record under investigation (e.g. Carré et al. 2005b; Colonese et al. 2012; Hallmann et al. 2008) to generate fine-grained local palaeoenvironmental data (Ulm 2013). Environmental proxy data derived from archaeological assemblages allow researchers to extend historical instrumental records and explore the effects of environment and climate on aspects of human behaviour at intra-regional levels (Andrus 2011). Yet, many archaeological studies instead rely upon regionally scaled or geographically removed environmental reconstructions (e.g. Brockwell et al. 2013; Stephens et al. 2008), potentially subjecting associated interpretations to latent biases and inaccuracies.

A growing body of research is exploring the efficacy of utilising accreting skeletal tissues to produce high-resolution reconstructions and interpretations of palaeoenvironmental conditions and their effects on past human behaviours (see Burchell et al. 2013c; Carré et al. 2005b; Eerkens et al. 2013; Fenger et al. 2007; Hallmann et al. 2008; Hufthammer et al. 2010; Jew et al. 2013; Mannino et al. 2003; Prendergast et al. 2013; Wurster and Patterson 2001 among others). Aspects of the ambient environment are recorded within the sequential growth structures of accreting skeletal tissues, preserving high-resolution time-series information (potentially reaching sub-daily scales) ideal for localised palaeoenvironmental reconstruction. Studies of a wide range of accreting skeletal tissues, including fish otoliths (Andrews et al. 2003; Hufthammer et al. 2010; Wurster and Patterson 2001; Wurster and Patterson 2003), teeth (Pike-Tay and Cosgrove 2002), coralline sponges (Böhm et al. 2000), some mammalian bone – including deer (Frankel et al. 2013) and wallaby (Pike-Tay and Cosgrove 2002) – and molluscan shell (Burchell et al. 2013b; Hallmann et al. 2009), show the rapidly expanding application of these techniques into a range of archaeological materials and investigations. Here we focus on one class of materials that is ubiquitous in coastal archaeological contexts – molluscan shell.

To access these archives researchers employ fine-scale data recovery methods, such as sclerochronology. Often referred to as the aquatic equivalent of dendrochronology (Andrus 2011), sclerochronology is the study of information preserved in the in the sequential growth structures of accretionary hard tissues (Andrus 2011; Gröcke and Gillikin 2008; Oschmann 2009). While the term was originally coined by Buddemeier et al. (1974) and Hudson et al. (1976) in reference to the study of density bands in stony head coral, its definition has since broadened to encompass a variety of physical and geochemical techniques (Oschmann 2009). This paper focuses on two commonly employed sclerochronological techniques – growth feature analysis and stable isotope analysis – as well as a complementary method – trace element analysis. Growth feature analysis is commonly used to assess macro- and microscopic physical variability in the growth regimes of accreting skeletal tissues (Burchell et al. 2013b; Burchell et al. 2013c; Claasen 1988), while stable isotope and trace element analyses allow the reconstruction of "daily", seasonal or longer-term environmental cycles as well as other facets of the palaeoenvironment (e.g. Carré et al. 2005b; Cohen et al. 1992; Hallmann et al. 2008; Kennett and Voorhies 1995). Similarly, growth feature analysis and stable isotope analysis are increasingly applied to address archaeological questions relating to population dynamics including occupation periodicity and seasonality, mobility and demography (e.g. Burchell et al. 2013c; Deith 1983; Eerkens et al. 2013; Jones et al. 2005; Shackleton 1973), allowing increasingly nuanced interpretations of human-environment relationships. Preliminary studies indicate that trace element analysis may provide similar information (e.g. Freitas et al. 2005; Surge and Walker 2006), however further research is required to verify these findings (Carré et al. 2006; Freitas et al. 2005). While these methods are routinely implemented by researchers from a variety of disciplines (e.g. geosciences, fisheries and environmental sciences) coastal archaeologists are increasingly utilising this toolset in a variety of geographic locations and climate zones, with special issues or sections of *The Journal of Island and Coastal Archaeology* (see West 2013) and other peer-reviewed sources providing a dedicated discourse (Figure 1).

While the potential of employing proxies derived from archaeological deposits to assist with palaeoenvironmental reconstruction has long been recognised (e.g. Aguirre et al. 1998; Kirby et al. 1998), a number of methodological challenges continue to hinder accurate interpretation. The use of poorly calibrated or inappropriate proxies and broad extrapolations of localised results can severely limit the accuracy and validity of analysis and associated data. While robust sampling strategies and careful selection of proxies can assist in minimising the effects of these issues, the literature indicates that many studies are unsuccessful in incorporating such measures (e.g. Brockwell et al. 2013; Kennett and Voorhies 1996; Lightfoot and Cerrato 1988; Stephens et al. 2008). Thus, many data sets are potentially poorly calibrated or do not operate at the resolution required for meaningful interpretation.

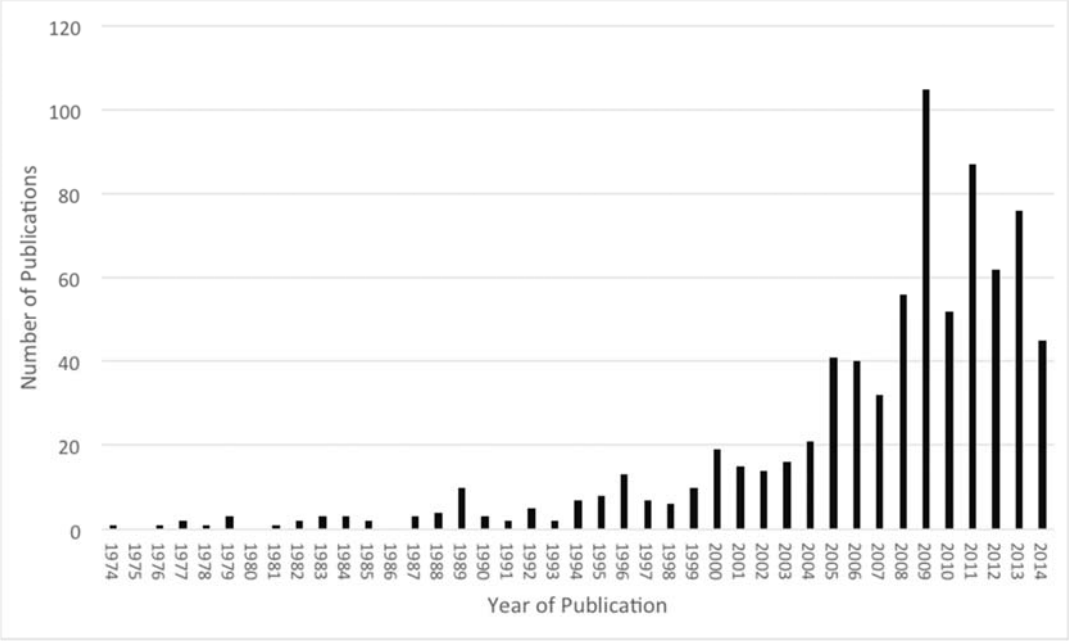

Figure 1. Yearly number of publications from a variety of disciplines (including archaeology, geosciences, fisheries and environmental sciences) from peer-reviewed sources with the search term sclerochron* between 1974 and 2014. Data from Web of Science database search on 03 November 2014.

This paper examines previous attempts to characterise the palaeoenvironment and explore human-environment interactions through archaeological proxies, highlighting methodological advantages and limitations of sclerochronology. Provided a variety of methodological and practical criteria are met, we posit that applying sclerochronological techniques to the accreting skeletal tissues of molluscan shell provides a means to accurately and meaningfully interpret past human-environment relationships.

2. Understanding Human-Environment Relationships through Environmental Proxies

Environments play a vital role in the everyday lives of humans, providing the space and conditions within which populations operate. As such, changes in environmental conditions act as fundamental structuring agents on human lifeways, potentially effecting behaviour on a variety of scales (Rowland 1983; Rowland 1999; Ulm 2013), making understandings of the environments in which past populations lived essential to addressing many fundamental archaeological questions. However, it must be stressed that environmental conditions cannot be viewed as the only factor driving change in human behaviour as this notion discounts the complexities of culture and society (Rowland 1999). Conversely, placing too great an emphasis on social or cultural mechanisms as the catalyst for behavioural change disregards the importance of environmental inputs (contrary to arguments by authors including Barker 1996; David and Lourandos 1999; Lourandos 1997; Lourandos and Ross 1994; Walters 1992). Environment can instead be viewed as a framework within which culture operates (Faulkner 2013; Rowland 1999; Veth et al. 2000), forming a continuous dialogue between both sides rather than a dichotomous relationship where humans adapt *in response* to environmental conditions (Anderson 2009; Balée 1998:14). Thus, the interface between culture and environment fundamentally influences human decision-making and is of most interest to archaeologists. By framing human-environment relationships against this backdrop, researchers are able to characterise complex changes in behaviour, culture and economy (Faulkner 2013; Rowland 1999).

Environmental proxies allow researchers in many disciplines to extend instrumental records, characterise past environments and identify relationships between environmental conditions and human behaviour (e.g. Burchell et al. 2013c; Carré et al. 2005b; Chivas et al. 2001; Cornu et al. 1993; Hassan et al. 2012; Kennett and Voorhies 1996; Reeves et al. 2007; Shulmeister and Lees 1995). A wide array of *in situ* proxies (e.g. ice cores, speleothems, tree rings and sediments) are employed to gather information regarding aspects of the palaeoenvironment – including temperature, salinity, biotic responses to climate change, ENSO cycles and broad-scale climate variations (Birks et al. 2010; Böhm et al. 2000; Carré et al. 2005b; Cohen et al. 1992; Marwick and Gagan 2011; Prendergast et al. 2013; Taft 2013). While these proxies provide vital information for archaeologists interested in exploring temporally or spatially expansive trends (e.g. Smith et al. 2008; Turney and Hobbs 2006; Williams et al. 2010), increases in resolution require finer time resolved data. As such, archaeologists must instead rely on environmental analogues recovered from archaeological deposits – including molluscan shell, fish otoliths, terrestrial fauna remains, environmentally sensitive flora and charcoal (see Faulkner 2008; Frankel et al. 2013; Hallmann et al. 2013; Hufthammer et al. 2010; Pike-Tay and Cosgrove 2002). Unlike other environmental proxies, those extracted from archaeological settings provide direct linkages between environment and human activity (Prendergast and Stevens in press), allowing researchers to locally contextualise environmental records and approach archaeological questions regarding human decision-making processes.

Employing proxies derived from the archaeological record requires researchers to overcome a variety of practical and methodological limitations. While a wide selection of proxies are often available, their viability or usefulness can be restricted by a series of research-specific factors. Geographic location, climate, temporal and spatial scale, required resolution, type of information sought and issues related to preservation and deposition/taphonomy all potentially affect the accuracy and validity of interpretations made from proxy sources. Failure to consider these can introduce latent inaccuracies and biases into data sets. For instance, the extrapolation of high-resolution proxies which document localised environmental conditions at short timescales (sub-daily or seasonally) into broader temporal and geographical contexts can diminish the accuracy of interpretations (e.g. Brockwell et al. 2013). Detail is lost as localised patterns are homogenised into generic overarching explanations (for further discussion on the impacts of normative methodologies see Claassen 1991). Thus, in order to mitigate the impacts of these potential issues archaeologists must carefully select proxies that fit criteria specific to the aims of their research.

While the selection of appropriate material assists in ameliorating interpretations from erroneous data (Burchell et al. 2013b; Hallmann et al. 2013; Kingston et al. 2008), a nuanced understanding of the biology, ecology and potential limitations associated with the chosen proxy is essential in generating meaningful and robustly interpretable results. Archaeologists are increasingly employing calcium carbonates derived from accreting skeletal tissues as a staple palaeoenvironmental proxy. The robust nature and ubiquity of accreting skeletal tissues in archaeological depositions has allowed their efficacy as palaeoenvironmental proxies to be rigorously examined (e.g. Campana 1999; Monks 1981:193-211; Schöne 2008). Most importantly, the effects of environmental conditions on chemical and physical elements of accreting skeletal tissues (e.g. growth regimes and isotopic fractionation) have been thoroughly tested and characterised in both laboratory and field settings (e.g. Epstein et al. 1953; Foster et al. 2009; Grossman and Ku 1986; Kim and O'Neil 1997; Turner 1982). This provides researchers with a comprehensive methodological foundation that accurately describes interactions between accreting skeletal tissues and the environment.

3. Growth Features, Stable Isotopes and Trace Elements: High-Resolution Archaeological Tools

Growth feature and stable isotope analysis are widely used to access and interpret fine-grained information archived within the growth structures of accreting skeletal tissues to construct temporally sensitive data profiles (Álvarez et al. 2011; Andrus 2011; Jones 1983). While the foundations of these techniques have been in place since the early 1950's (Epstein et al. 1951; Epstein et al. 1953; Haskin 1954; McCrea 1950; Orton 1923; Urey 1947; Urey et al. 1951), sclerochronological protocols have only recently been widely applied to archaeological contexts (e.g. Burchell et al. 2013a; Hallmann et al. 2013; Schöne 2013; among

others). Conversely, trace element analysis is a relatively new technique, requiring further research to better understand the fundamental links between trace element composition in molluscan shell and environmental conditions (Carré et al. 2006; Freitas et al. 2005). Recent advances in sampling and analysis have led to significant increases in resolution and accuracy (Merritt and Hayes 1994; Spötl and Mattey 2006; Wurster et al. 1999) encouraging increasingly routine application of these techniques. Thus, the efficacy of utilising growth features, stable isotopes and trace elements in a range of climatic and environmental settings is under constant review, with results developing into an increasingly nuanced theoretical foundation that highlights and addresses discipline specific challenges (Goodwin et al. 2004; Schöne 2008).

The fundamental concepts of sclerochronology rely on aspects of local environmental conditions being archived sequentially within the biogenetic carbonates of skeletal structures (Epstein et al. 1953; McCrea 1950; Pannella and Copeland 1968; Rhoads and Pannella 1970; Urey 1947). Thus, underlying requirements for target material are that carbonates precipitate in equilibrium with the ambient environment and are deposited in periodic growth layers to allow precise temporal alignment of the chemical record with the environmental changes being recorded (Schöne and Surge 2012). While these attributes have been observed in a variety of skeletal material found within the archaeological record, molluscan shell (particularly bivalves) is most frequently utilised owing to its broad geographic distribution throughout a diverse range of environments and climates, sensitivity to environmental change, sequential growth patterning, high potential for preservation and extensive history of research (Bailey et al. 1983; Clark 1974; Rhoads and Lutz 1980).

During shell growth the chemical and physical interactions between precipitating calcium carbonates and the ambient environment imprint molluscan skeletal tissues with representations of environmental and hydrological cycles (Eisma et al. 1976; Grossman and Ku 1986; Rhoads and Lutz 1980). While these cycles primarily reflect regular occurrences such as tides or seasonal variation, irregular events including extreme weather or physiological change can also be recorded (Andrus and Rich 2008; Jones et al. 2005). Therefore, employing high-resolution data recovery techniques, such as sclerochronology, to systematically analyse molluscan shell allows the generation of detailed chronological life histories and reconstructions of the ambient environment. The application of these methods to archaeological assemblages provides the opportunity to address broader questions regarding palaeoenvironments, climate change and their effects on human behaviour (Burchell et al. 2013b; Clark 1974; Schöne and Surge 2005).

3.1 Growth Feature Analysis

Researchers use growth patterns to understand life history traits of target taxa, assist in palaeoenvironmental reconstruction and provide temporal context for higher-resolution analysis (Andrus and Rich 2008; Hallmann 2011). Shifting environmental and climatic conditions as well as biological processes act as fundamental controls on the growth regimes of molluscs. Variations in water temperature and mixing can significantly alter shell precipitation rates, a process which manifests as discrete growth banding (Clark 1974; Lutz and Rhoads 1977; Rhoads and Lutz 1980). Biological (circadian) clocks, operating at regular rhythms, ensure that growth banding continues to be produced when molluscs are not exposed to significant environmental change (Richardson 1987; Richardson 1988; Schöne 2008; Schöne 2013). Visible on the exterior, hinge and in cross-section, growth banding is comprised of two basic structures: lines and increments (Figure 2). Lines represent periods of growth slowdown brought about by environmental stressors that exceed the tolerances of the mollusc. It must be noted that cases of total growth cessation are not evidenced within the skeletal structures of molluscan shells as carbonates are not precipitated during this time (Schöne 2008). Increments correspond to periods of normal or accelerated growth, often correlating to more optimal environmental conditions. As the deposition of banding is maintained and regulated by biological clocks and recurring environmental events, the periodicity of which range between diurnal tidal cycles and annual cycles (Andrus 2011; Clark 1974; Deith 1983; Richardson 1987; Richardson 1988; Schöne 2008; Schöne 2013), researchers are able to interpret growth features as physical expressions of time, provided the drivers of band formation are known.

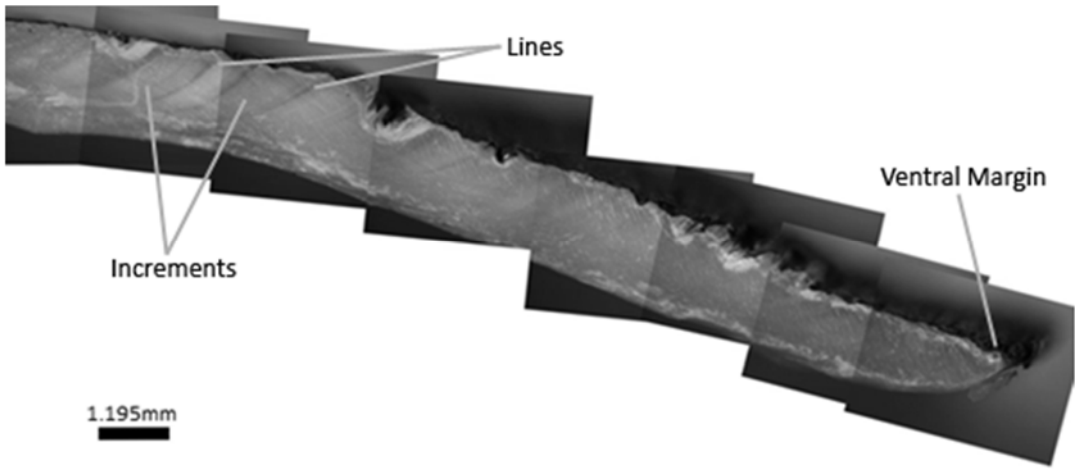

Figure 2. Growth lines and increments from a live-collected *Polymesoda coaxans* (image adapted from Hinton 2012:33).

Early research concentrated on macroscopically visible growth features, applying visual and metric analysis (a process Deith (1983:425) termed 'growth-line analysis') to infer season of death and age of individuals (e.g. Coutts 1970; Deith 1983; Lightfoot and Cerrato 1988; Lightfoot and Cerrato 1989). Growth-line analysis protocols assumed that shell growth remained consistent between taxa as well as through time and space, except during the winter when lowered temperatures would cause growth to slow forming a distinctive 'winter' growth line (Monks 1981; Rhoads and Pannella 1970). Pairs consisting of a line and an increment were used to delineate annual cycles allowing counts to be employed as an estimate of age. Season of death was determined by measuring and dividing full years of growth into equal segments representing 'seasons' (Monks and Johnson 1993). As the final pair is often truncated by the death of the organism, the size disparity between it and the previous year's growth was used to align the time of death to specific 'seasons' (Monks and Johnson 1993).

Recent high-resolution studies of shell growth regimes have revealed that growth-line analysis is overly simplistic as environmental stimuli do not produce uniform effects in molluscan growth, with distinct variability detected between taxa and through time and space (Bernstein 1990; Henry and Cerrato 2007; Jones and Quitmyer 1996). The wide variety of ecological zones inhabited by molluscs requires further consideration of interactions between shellfish and environmental conditions. For instance, tropical zones frequently experience more stable temperatures than temperate areas, meaning changes in growth may be attributable to other variables, including hydrological changes or availability of nutrients (Gillikin et al. 2005a; Kennett and Voorhies 1996; Stephens and Rose 2005). Many researchers now consider the visual inspection of macroscopic growth features alone inadequate to accurately determine season of death or age (see Andrus 2011). Consequently, the study of macroscopic growth features has been repurposed with researchers employing them in conjunction with complementary finer-scaled techniques to gain temporal and spatial understandings of high-resolution analyses. As each growth structure represents a discrete period of time, growth features provide a physical framework that allows archaeologists to study isolated time-intervals at a variety of scales. Thus, researchers have begun employing this method when undertaking carbonate sampling for stable isotope analysis, allowing individual samples to be contextualised within a broader temporal setting while maintaining spatial control (e.g. Andrus and Rich 2008; Burchell et al. 2013a; Burchell et al. 2013b; Elliot et al. 2003).

Researchers have begun testing the efficacy of utilising micro-growth features (Figure 3) to increase the accuracy and resolution of archaeological interpretation (e.g. Burchell et al. 2013a; Burchell et al. 2013b; Custer and Doms 1990; Hallmann et al. 2009; Schöne et al. 2005b). Formed in a similar fashion to

macroscopic banding, micro-growth features correspond to significantly shorter periods of time. While the timing and periodicity of micro-growth formation is primarily regulated by the biological clock of molluscs, tidal regimes may also be reflected (Richardson 1987; Richardson 1988; Schöne 2008; Schöne 2013). When some intertidal molluscs are submerged during high tide shell precipitation is accelerated and a micro-growth increment is produced (Goodwin et al. 2001), as the tide subsides and the shellfish is aerially exposed growth slows forming a micro-growth line (Evans 1972). This process also causes physical discrepancies between tides with variable amplitudes (primarily neap and spring) owing to the amount of time the mollusc is exposed to the corresponding condition (Hallmann et al. 2009). Neap tides, which see reduced tidal variation, produce much broader increments and less defined micro-growth lines while the increased tidal amplitude associated with spring or king tides generate highly defined micro-growth lines and narrow increments (Hallmann et al. 2009). Provided attributes of the local tidal regime and target species are known, groupings of micro-growth features can be utilised as an exceptionally high-resolution expression of time allowing researchers to precisely characterise growth rates, differentiate regular and irregular growth stoppages, and when the date/time of capture are known, accurately align growth features to calendar dates (Burchell et al. 2013b; Carré et al. 2009; Hallmann et al. 2009).

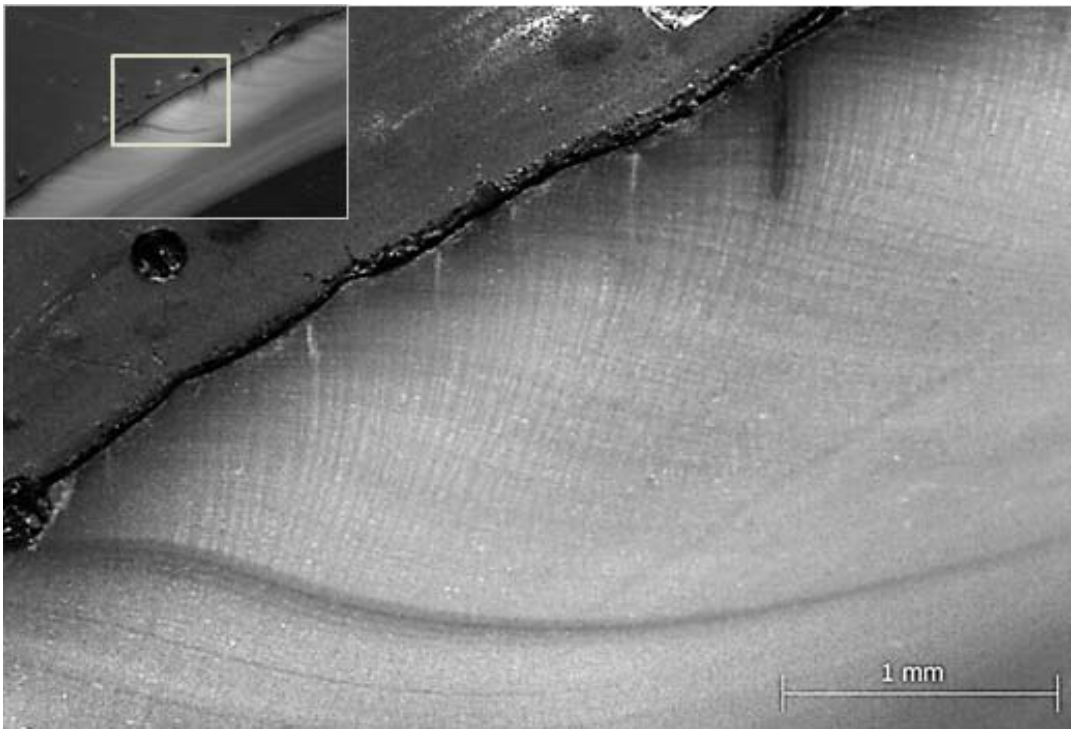

Figure 3 Micro-growth features in comparison to a larger macroscopic growth-line found in a *Polymesoda coaxans* valve live-collected from Bentinck Island, Gulf of Carpentaria, Australia.

3.2 Stable Isotope Analysis

Stable isotope analysis applies mass spectrometry to measure relative abundances of isotopes in carbonate samples extracted from accreting skeletal tissues. While both stable and unstable (radiogenic) isotopes are detected, this paper focuses on stable isotope analysis as they are of most use to palaeoenvironmental reconstruction. The resulting isotopic signals provide detailed information about modern and past environments, allowing archaeologists to approach higher-order interpretations of human-environment relationships. This method assumes that during growth shell calcium carbonates are imprinted with a chemical depiction of environmental conditions owing to changes in rates of isotopic fractionation and the isotopic value of ambient water. Relative abundances of isotopes are expressed in delta notation (e.g. $\delta^{18}O$), a ratio of heavy isotopes (e.g. ^{18}O) to light isotopes (e.g. ^{16}O) measured against international reporting

standards such as the Vienna Pee Dee Belemnite (VPDB). Due to the extremely small differences being described, delta values are reported as parts per mil (‰) variations from a standard with a defined value of 0‰. More positive values (e.g. $\delta^{18}O = +3.1‰$) signify a higher ratio of heavier isotopes, while more negative values (e.g. $\delta^{18}O = -2.3‰$) indicate a higher ratio of lighter isotopes (Fry 2006:22-23). The generation of delta values requires isotopic content of the material in question to be measured against an appropriate standard using equation (1):

$$\delta(‰) = [(R_{sample} / R_{standard} - 1)] \times 1000 \quad (1)$$

Where *R* is the ratio of the heavy-to-light isotopes, R_{sample} is the isotopic ratio of the sample being assessed, and $R_{standard}$ is the international standard (Epstein et al. 1953).

3.2.1 Oxygen Isotope Values

Studies attempting to characterise past environments and population dynamics typically focus on analysing the relationship of isotopic values of oxygen in molluscan calcium carbonates ($\delta^{18}O_{shell}$) to environmental variation (Figure 4). As a majority of studied bivalves precipitate their shells near to oxygen isotopic equilibrium with ambient conditions (Mook and Vogel 1968), water temperature and composition act as primary controls on oxygen isotope fractionation between environment and shell. Variations in either are recorded chemically within the shell matrix. Temperature shares an inverse relationship with $\delta^{18}O_{shell}$, i.e. more negative values represent higher temperatures and vice versa (Epstein et al. 1953; Urey 1947), owing to a negative correlation between temperature and the fractionation process whereby every 4-5°C of temperature variation causes an approximately 1‰ change in $\delta^{18}O_{shell}$ values (Grossman and Ku 1986; Shackleton 1973). Conversely, the $\delta^{18}O$ of water – a function of evaporation, precipitation, terrestrial runoff and water mixing (Bemis and Geary 1996; Dansgaard 1964) – shares a positive correlation with $\delta^{18}O_{shell}$. The process of evaporation or input of marine waters increases the $\delta^{18}O$ value of source water while simultaneously increasing the salinity of the environment. Conversely, influxes of freshwater, for example precipitation or run-off, simultaneously lower salinity and the $\delta^{18}O$ value of source water. However, it must be noted that while salinity and $\delta^{18}O$ frequently co-vary they are not directly linked and therefore interpreting any perceived association must be approached with caution.

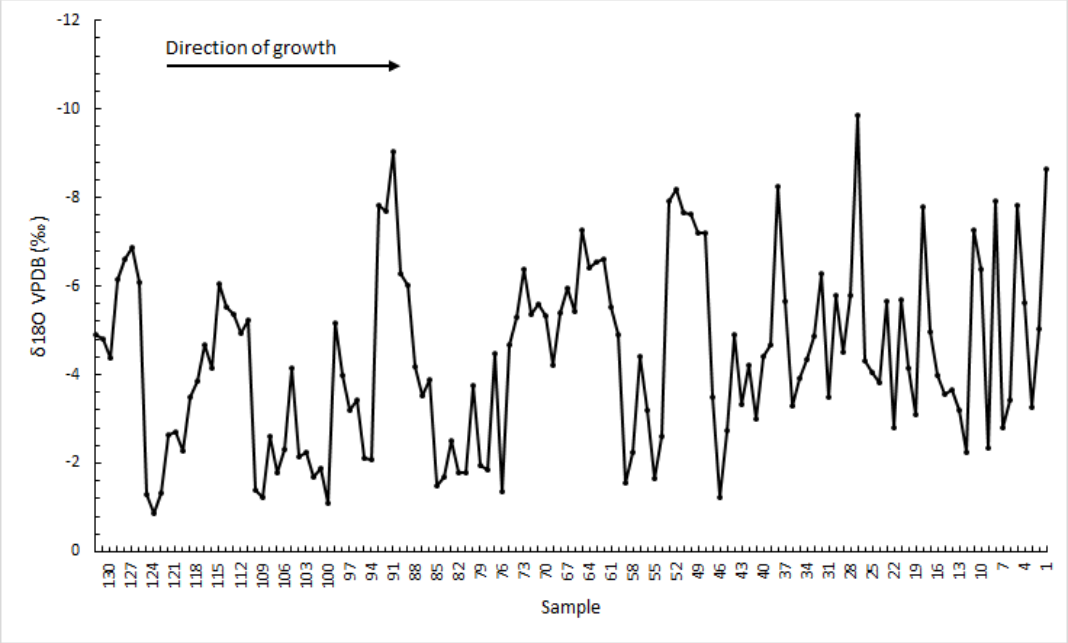

Figure 4. Oxygen isotope profiles of molluscs can provide insight into the environmental conditions experienced by the organism throughout its lifetime. Note the regular oscillations occurring throughout ontogeny of this Polymesoda coaxans valve. These can become increasingly difficult to interpret owing to increased crowding of growth structures with age, as seen in samples 1 – 12 of this specimen (figure adapted from Hinton in prep).

In archaeological settings, variations in $\delta^{18}O_{shell}$ values are most commonly used to explore timing and periodicity of occupation. Pioneered by Shackleton (1973), this method takes advantage of the well-documented correlations between relative oxygen isotope abundances, temperature and water constitution to provide precise seasonal determinations (Böhm et al. 2000; Keith et al. 1964; McCrea 1950). As molluscs precipitate shell carbonate sequentially, the outer-most growth features formed most recently. Thus, oxygen isotope values procured from these structures potentially reflect the ambient environment at the time of the organism's death. These signals can be aligned to seasons via comparisons with modern control samples, generated using data from modern specimens, water samples and observed environmental records, allowing an accurate determination of season of death/capture (Andrus 2011; Milner 2001). When applied throughout archaeological assemblages high-resolution histories of occupation can be generated, in some cases allowing researchers to approach higher-order interpretations of population dynamics including mobility and demography (Burchell et al. 2013a; Burchell et al. 2013b; Burchell et al. 2013c; McManus et al. 2013; Thompson and Andrus 2013). Mounting interest in increased archaeological resolution has led many researchers to apply these methods in order to gain nuanced understandings of human-environment interactions (e.g. Hallmann et al. 2013; Kennett and Voorhies 1995; Kennett and Voorhies 1996; Mannino et al. 2011; among others), such as exploring links between intensity of shellfish harvest and seasonal shifts in environmental conditions (see Burchell et al. 2013b). It should be noted that many of these studies have been undertaken in temperate zones where significant oscillations in temperature are common between seasons (Kennett and Voorhies 1996). Tropical regions are subject to lower amplitude shifts in temperature, thus its impact on mollusc growth and chemistry is potentially lessened. However, other environmental cycles that impact molluscan calcium carbonates, such as influxes of freshwater from monsoonal rainfall, frequently coincide with seasonal change in the tropics, generating an equally practical chemical record (see Hinton 2012; Kennett and Voorhies 1995; Kennett and Voorhies 1996; Stephens et al. 2008).

Due to the relationship between oxygen fractionation and temperature (Grossman and Ku (1986) report a 1‰ change in $\delta^{18}O_{shell}$ reliably equating to a 4-5˚C temperature variation), $\delta^{18}O$ values derived from carbonate are widely used as part of palaeotemperature modelling (e.g. Cornu et al. 1993; Goodwin et al. 2001; Prendergast et al. 2013; Urey et al. 1951). Carbonate samples are collected through shell ontogeny and resulting isotopic values are transformed into temperature estimates using palaeotemperature equations (see Grossman and Ku 1986). The relationship between temperature and $\delta^{18}O_{shell}$ values differ between calcium carbonate polymorphs (Böhm et al. 2000; Epstein et al. 1953; Grossman and Ku 1986). A majority of molluscan shells are formed from calcite, aragonite or a combination of both (Claassen 1998:22; see Nehrke et al. 2012 for an example of molluscs and incorporate a third carbonate polymorph, vaterite), thus care must be taken to accurately identify the polymorph being analysed prior to applying equations (see Section 4.2 for further discussion). Variations in temperature can then be characterised over the life span of the mollusc.

This technique has been applied to long-lived organisms, such as corals or deep sea shellfish, to produce extended climate records (Cohen and Tyson 1995; Helama and Hood 2011; Schöne 2013), while shorter-lived taxa, such as foraminifera and molluscs, have been used to produce temporally constrained temperature estimates (Fenger et al. 2007; Jones and Kennett 1999; Mannino et al. 2008; Prendergast et al. 2013). Similar to dendrochronology, long-lived species can be used to generate master chronologies to extend records beyond the lifetime of a single organism via cross-dating techniques (e.g. Butler et al. 2010; Jones et al. 1989; Marchitto et al. 2000; Witbaard et al. 1997). However, equations associated with palaeotemperature require precise knowledge of past local $\delta^{18}O_{water}$ values to produce accurate reconstructions (see Epstein et al. 1953; Grossman and Ku 1986; McCrea 1950). This proves problematic within archaeological contexts as data relating to ancient water sources are often unattainable, meaning the composition must be assumed or estimated, introducing an additional source of uncertainty and inaccuracy. Researchers are attempting to address this issue by exploring the efficacy of alternative means of temperature reconstruction such as clumped isotope analysis (see Ghosh et al. 2006; Ghosh et al. 2007; Schöne 2013). However, the slope of the associated regression line is very shallow, thus requiring large quantities of carbonate (~10 mg) to gain relatively accurate temperature determinations making it impractical in many situations.

As temperature and the isotopic value of ambient water are primary determinants of the oxygen isotope composition of calcium carbonates, the value of $\delta^{18}O_{shell}$ can vary as a function of changes in temperature, influxes of freshwater or both. In regions with environmental shifts dominated by either temperature or water constitution, establishing causality between isotopic signals from environment and carbonate is relatively simple. However, in areas subject to the dual influences of temperature and variable water mixing (e.g. estuarine systems or some tropical regions) disentangling these effects can be difficult, particularly with regards to archaeological material where environmental data are not independently known (Andrus and Thompson 2012; Culleton et al. 2009). Some authors have gone so far as to reject all material from such habitats, stating that their inherent complexities make them too difficult or inaccurate to analyse (e.g. Godfrey 1988; Milner 2001). However, a growing body of research has demonstrated the efficacy of utilising oxygen isotopes in areas with more complex temperature-water constitution relationships, particularly when supplemented with high-resolution growth feature analysis, comparative modern baselines, or further stable isotope analysis (Andrus and Crowe 2000; Burchell et al. 2013a; Burchell et al. 2013b; Burchell et al. 2013c; Jones et al. 2004; Jones et al. 2005; Kennett and Voorhies 1995; Kirby et al. 1998).

3.2.2 Carbon Isotope Values

Stable carbon isotopes (^{12}C and ^{13}C; $\delta^{13}C$) are also useful to archaeological investigation. However, the parameters controlling carbon isotope fractionation are more complex than for oxygen, and as such are not employed as readily (Kirby et al. 1998; Wefer and Killingley 1980). Functions dictating carbon isotope fractionation cannot be attributed to a single control, such as temperature or water mixing. Instead the $\delta^{13}C$ value is derived from a mixture of environmental dissolved inorganic carbon (DIC) and metabolic CO_2, both of which fluctuate depending on season, community photosynthesis, respiration and metabolism (Kennett and Voorhies 1995; Kirby et al. 1998; McConnaughey and Gillikin 2008). However, despite the

complexities, cautious application of carbon isotopes can provide valuable information concerning biological cycling, vegetation type, water source, and/or diet (Geary et al. 1992; McConnaughey and Gillikin 2008; Petchey et al. 2013; Petchey et al. 2012; Stephens et al. 2008).

A majority of the difficulties associated with interpreting carbon isotope signals are derived from chemical and physiological processes which occur during carbonate precipitation. While thought to be relatively uncommon in molluscan shell (McConnaughey and Gillikin 2008), kinetic effects can cause calcium carbonates to precipitate in disequilibrium with the ambient environment – meaning isotope values no longer reflect surrounding conditions. This is primarily a product of discrimination against ^{13}C (or ^{18}O) isotopes at higher rates of carbonate precipitation (McConnaughey 1989; Schöne and Surge 2012), making it likely only in exceptionally fast growing sections of shell.

Molluscan $\delta^{13}C_{shell}$ values largely reflect ambient $\delta^{13}C$ values, providing chemical information regarding the environment in which the mollusc lived and grew (McConnaughey and Gillikin 2008; Schöne 2008). Some debate surrounds the amount of metabolic CO_2 incorporated into molluscan shell, particularly in detritus feeders and gastropods where large percentages (up to 85%) of shell carbonate can originate from metabolic carbon (Tanaka et al. 1986). This suggests that $\delta^{13}C_{shell}$ signals can be further complicated by carbon derived from diet, causing shell carbonate to enter a state of disequilibrium with the external environment (McConnaughey 1989). For this reason few archaeological studies utilise $\delta^{13}C$ values derived from gastropods as a source of information. Instead, researchers focus on filter-feeding bivalves that only incorporate a small fraction of respired CO_2 into their skeletal structures (Gillikin et al. 2006; McConnaughey and Gillikin 2008; Mook 1971; Mook and Vogel 1968). However, it must be remembered that even though the bulk of carbon may be derived from DIC, $\delta^{13}C_{shell}$ values are potentially altered by a variety of other stimuli, including environmental CO_2/O_2 ratios, DIC content, and physiology (see McConnaughey and Gillikin 2008).

In coastal contexts $\delta^{13}C$ values can assist in tracking shifts in salinity and water mixing. The $\delta^{13}C$ value of fluvial DIC, typically less than -8‰ although this can be highly variable (Kennett and Voorhies 1995), is often distinctly lower than marine DIC, typically -2‰ (Kennett and Voorhies 1995), due largely to the decomposition of terrestrial plants and soil processes (Keith et al. 1964; Kennett and Voorhies 1995; McConnaughey and Gillikin 2008). Thus, $\delta^{13}C$ values potentially replicate the mixture of marine and fluvial DIC in a positive correlation, i.e. the greater the marine influence the more positive the $\delta^{13}C$ value (Gillikin et al. 2006; Mook and Vogel 1968). When used in conjunction with $\delta^{18}O$ a more precise depiction of mixing regimes is established (McConnaughey and Gillikin 2008), assisting researchers in delineating the dual effects of water constitution and temperature and increasing the accuracy of seasonal determinations (Andrus and Crowe 2000).

3.3 Trace Element Analysis

Trace element analysis, a parallel method of geochemical assay, has recently begun to see increasing use within archaeological research. While a variety of analytical techniques can be utilised in conjunction with numerous materials – for example the study of corals using ion probes (e.g. Allison 1996; Hart and Cohen 1996) and laser inductively coupled plasma mass spectrometry (ICP-MS) (e.g. Sinclair et al. 1998) – this paper focuses on the analysis of molluscan calcium carbonate samples as it is directly comparable to the previously discussed procedures for stable isotope analysis. Like stable oxygen and carbon isotopes, trace element abundance in calcium carbonates is thought to be controlled by physical and chemical attributes of ambient water. By analysing ratios of element/Ca of archaeological calcium carbonates, researchers can potentially reconstruct aspects of past environments during calcification (particularly temperature) and track oscillations in environmental conditions (e.g. Beck et al. 1992; Freitas et al. 2005; Gillikin et al. 2005b; Surge and Walker 2006). However, controls governing trace element incorporation into biogenic carbonates

are not as well understood (Carré et al. 2006; Freitas et al. 2005), necessitating further research into the effects of environmental factors such as temperature, salinity and pH.

A range of trace elements are used in conjunction with a wide variety of organisms exhibiting biogenic carbonates, however studies focused on molluscan shell most commonly employ ratios of magnesium and/or strontium to calcium (Mg/Ca and Sr/Ca). The partitioning of Mg/Ca between solid and liquid phases is thought to be temperature-dependent (Mucci 1987) and thus potentially tracks changes in ambient temperature acting as a palaeothermometer (Klein et al. 1996). However, correlations between temperature and Mg/Ca can be weak with temperature reconstructions requiring precise knowledge of the environment during shell growth (Surge and Lohmann 2008), making Mg/Ca only suitable as an auxiliary proxy. Ratios of Sr/Ca have also been explored as a proxy for palaeotemperature (Freitas et al. 2006), however it was found that correlations were generally weak or inconsistent. Instead, it has been suggested that growth rates may have a substantial influence on Sr/Ca ratios in some molluscan taxa (Carré et al. 2006) or that salinity may impact strontium levels similar to fish otoliths (see Elsdon and Gillanders 2005).

Conversely, other studies have reported less consistent results. Gillikin et al. (2005b) noted a minimal positive correlation between Sr/Ca and temperature in the molluscan taxa *Saxidomus giganteus* and *Mercenaria mercenaria*, however these were too weak to act as a temperature proxy. Similarly, Freitas et al. (2005) found no link between Sr/Ca and temperature in *Pinna nobilus*. Both studies suggest that these findings resulted from biological and metabolic controls having a greater influence on Sr/Ca ratios than temperature. Ontogenetic changes in physiology have also been suggested as a cause of ambiguous results in Mg/Ca ratio-temperature correlations with Schöne et al. (2011) indicating that physiology can play a key role in element partitioning. Such results indicate that there is likely a strong species dependent component to the uptake and partitioning of trace elements within molluscan shell (Gillikin et al. 2005b; Schöne et al. 2011) and that like stable isotope analysis, trace element analysis requires a detailed understanding of target taxa physiology as well as local environmental and hydrological conditions.

4. Challenges Associated with Applying Sclerochronology to Archaeological Contexts

A number of methodological and discipline-specific challenges face archaeologists who seek to integrate sclerochronological analyses of molluscs into methodological frameworks. Not only is it essential to possess a sophisticated understanding of the techniques being employed but also of localised environmental cycles and conditions, taxa and location-specific relationships between growth and environmental conditions, the use of appropriate sampling procedures and methods to maintain temporal and spatial control and the effects of post-depositional alteration on the accuracy and validity of data.

To contextualise these challenges within a 'real world' research setting, two recent studies investigating palaeoenvironmental conditions in areas exhibiting complex environmental and hydrological cycles are employed below as examples where appropriate. Both studies examine a combination of modern and archaeological material to characterise environmental conditions in regions affected by strong monsoonal cycles. Hinton (2012) explores the efficacy of archaeological *Polymesoda* (*Gelonia*) *coaxans* (Gmelin, 1971) as palaeoenvironmental proxies for the South Wellesley Islands, Gulf of Carpentaria, Australia, while Stephens et al. (2008) utilises *P. coaxans* (although it is referred to by Stephens et al. (2008) by its synonym *Gelonia erosa*) as a palaeoenvironmental proxy for the area surrounding the Great Cave of Niah in Borneo, Malaysia.

4.1 Understanding Localised Environmental Impacts on Shellfish

The phylum Mollusca is highly diverse, consisting of approximately 85,000 extant species that inhabit a wide variety of geographic locations and climate zones. Such diversity makes it likely that shellfish taxa display unique responses to environmental stimuli owing to variable ecological tolerances. Thus, gaining detailed understandings of local environmental conditions and how they impact growth regimes and isotopic fractionation of relevant shellfish taxa is integral to accurate and meaningful sclerochronological analysis. While some differences may be obvious owing to distinct physiological or morphological attributes – e.g. bivalves/gastropods and filter/detritus feeders often produce divergent isotopic signals owing to dietary uptake – others are less so.

Recent high-resolution studies of molluscan growth regimes have characterised intra-species differences in rates of shell precipitation through time and space, demonstrating that oscillations in temperature and water composition can have inconsistent effects on the growth regimes of geographically distinct populations (Bernstein 1990; Henry and Cerrato 2007; Jones and Quitmyer 1996). Moreover, Hallmann et al. (2008) and Gillikin et al. (2005a) have demonstrated that individuals from the same population can produce variable isotopic signals, suggesting that this may be indicative of micro-habitat variability or differences in exchange rates between the outer extrapallial fluid (EPF) and the ambient water. Broader species-dependent growth variability has also been documented (e.g. Carré et al. 2005a). These findings add a further layer of complexity to the application of sclerochronology making it essential to treat shellfish populations independently as there is no guarantee as to how the ambient environment will have affected its molluscs in that environment. Thus, gaining understandings of local environments and establishing their effects on target shellfish taxa is integral to accurately interpreting sclerochronological data.

Establishing causality between modern environmental cycles and the growth regimes and isotopic fractionation of local shellfish populations also plays a vital role in securing meaningful interpretations derived from archaeological material. Although research employing sclerochronology to characterise palaeoenvironments and their effects on human behaviour is largely evidenced by isotopic signals and growth patterning of archaeological material, a contextual framework must be established to accurately parse this information (Andrus 2011; Jones and Quitmyer 1996; Mannino et al. 2003; Milner 2001). Without comparative data, results derived from archaeological material become difficult to accurately interpret.

Many studies have addressed these challenges through monitoring modern environments and shellfish populations in the vicinity of relevant archaeological settings. This not only provides researchers with an understanding of environmental cycles and their effect on target mollusc taxa but also establishes modern baselines that can be used to contextualise archaeological data. While environmental monitoring can take many forms, it often includes one or a combination of the following: water sampling for salinity, $\delta^{18}O$ and $\delta^{13}C_{DIC}$ values over discrete environmental cycles (tidal, seasonal, or annual) (Andrus and Rich 2008), live-collection and analysis of modern shellfish specimens (Hallmann et al. 2009), recording the ambient environment for temperature, tidal height and salinity (manually or via installing environmental sensors) (Andrus and Crowe 2000; Hallmann et al. 2008), and incorporating third-party observation records (Hinton 2012; Stephens et al. 2008). Ideally, all of these methods would be deployed to generate the most accurate picture of the local environment possible, however the remote nature of some archaeological research sites coupled with time and budget constraints often makes this difficult to achieve. These same restrictions affect the viability of long-term monitoring regimes making it difficult to characterise annual and inter-annual variability.

Archaeologists often address this by employing environmental modelling techniques in order to reconstruct aspects of the modern environment they were unable to directly observe. Equations which employ environmental data (such as sea surface temperature or rainfall records) in combination with isotopic signals from modern live-collected shell specimens or ambient water sources are commonly used to generate

predictive models of fractionation factors (Dettman et al. 1999; Grossman and Ku 1986), $\delta^{18}O_{shell}$ (Grossman and Ku 1986) and $\delta^{18}O_{water}$ values (Dettman et al. 1999). Researchers utilise predictive data to proxy missing environmental records (Hinton 2012) and cross-calibrate ambient conditions with physical and chemical characteristics of molluscan shell (Goodwin et al. 2001). Following collection, modelling and analysis, data taken from environmental, sclerochronological sources can be aligned to calendrical dates allowing causality between environmental controls, molluscan growth and isotopic signals to be characterised (e.g. Andrus and Crowe 2000; Hallmann et al. 2008). This serves as the comparative baseline analogue allowing the accurate interpretation of archaeological data.

Both Hinton (2012) and Stephens et al. (2008) undertook minimal environmental monitoring, instead relying upon third-party environmental records as the basis for modelling and comparisons. The monitoring that did occur took the form of nominal water sampling. Stephens et al. (2008) collected water samples from throughout the target river system over the period of a single day (see Stephens and Rose 2005 for a detailed account of sampling). While this affords geographically extensive knowledge of hydrological conditions throughout the region, it only provides a temporally limited snapshot. Furthermore, no comparisons are drawn between the isotopic signature of the river water and isotope values derived from shell carbonate – likely owing to the temporally restricted nature of the sampling regime. Instead, carbonate samples are matched to observed rainfall records, assuming the impacts of changing water constitution. While these records appear to mesh well, the accuracy of these results cannot be verified without direct evidence of *P. coaxans* response to localised changes in environmental conditions.

Hinton (2012:58) incorporated three water samples in order to 'provide a basic dry-season estuarine isotopic water composition value'. Two samples were taken at the collection site of modern *P. coaxans* at an interval of one month, the third was collected from the mouth of the adjacent creek system, Mirdidingi Creek, to provide an estimate of intra-creek variability. Akin to Stephens et al. (2008), the limited scope of sampling makes comparisons between environment and isotopic values of shell carbonate difficult. To overcome this Hinton (2012) undertook environmental modelling to estimate $\delta^{18}O_{water}$ values from observed environmental data, allowing the generation of a predictive $\delta^{18}O$ curve for comparison with isotopic profiles derived from modern specimens. While mathematical modelling does not provide data as accurate or nuanced as direct repeated water sampling, it does allow cautious interpretations of molluscan responses to environmental conditions. Hinton (2012) acknowledges that the limited water sampling may have impacted the scope of interpretation, recommending more intensive sampling for future research to afford increased accuracy and nuance in understandings of interactions between *P. coaxans* and local environmental conditions.

4.2 Countering Post-Depositional Alteration to Carbonate

Issues associated with post-depositional, taphonomic and site formation processes present significant challenges to all archaeological research. The interpretation of coastal sites in particular is often considered extremely complex owing to uncertainties relating to site preservation and post-depositional factors which may distort the archaeological record (Bird 1992; Bird 1995; O'Connor and Sullivan 1994; Rowland 1992; Rowland and Ulm 2012). While shell-rich features regularly provide excellent preservation conditions for constituent components – raised pH levels and layers of robust shell provide protection for delicate material (Andrus 2011) – damage or change stemming from post-depositional processes can lead to alteration of geochemical and physical elements of the shell itself effecting the accuracy of stable isotope analysis. Molluscan shell is particularly sensitive to modifications that lead to fragmentation, chemical alteration from contact with percolating ground waters, and recrystallisation (Bailey et al. 1983; Kennett and Voorhies 1996; Shackleton 1973).

Taphonomic processes can weaken the structural integrity of shell increasing its susceptibility to fracture and fragmentation. While a majority of archaeological shell deposits are subject to some degree of fragmentation, stable isotope studies have largely focused on analysing whole valves (e.g. Gillikin et al. 2005a; Mannino et

al. 2007; Shackleton 1973). This practice can severely limit specimen selection in assemblages which have undergone extensive fragmentation and may subsequently introduce biases and inaccuracies owing to under- or over-representation of taxa. The apparent reluctance to incorporate fragmented material likely stems from an uncertainty regarding the efficacy of this practice and concerns of isotopic inhomogeneity between varying portions of shell (Kingston et al. 2008). However, a growing body of research is exploring the value of fragmented material, suggesting that high-resolution data are attainable from fragmented assemblages provided caution is exercised when selecting sample material (Burchell et al. 2013a; Burchell et al. 2013b; Hallmann et al. 2013; Kingston et al. 2008).

Percolating ground waters, often originating from meteoric water, can chemically alter skeletal structures. Shell that has suffered structural damage or dissolution is particularly susceptible. Oxygen isotope exchange between the two can leave shell carbonate isotopically lighter owing to an abundance of ^{16}O in meteoric water (Bailey et al. 1983; Killingley 1981). Thus, isotopic signals derived from molluscan shell that come into contact with percolating ground water may no longer be representative of the environment in which it grew.

As previously discussed, crystalline forms of a majority of molluscan carbonate are comprised of aragonite, calcite or a combination of both (Claassen 1998:22). Exposure to temperatures of approximately 150°C will cause the organic matrix, on which the crystalline structure is built (Marin and Luquet 2004; Wilt et al. 2003), to break down and the crystal structure to expand (Collins 2012). As temperatures near 350°C aragonitic structures begin to invert or recrystallise into the more stable polymorph calcite, with complete conversion occurring at approximately 500°C (Collins 2012; Epstein et al. 1953). This change compromises the integrity of geochemical signals derived from the shell and significantly diminishes the accuracy of palaeoenvironmental determinations. As these temperatures are commonly exceeded by hunter-gatherer campfires (Robins and Stock 1990), pre-depositional human activities, such as cooking, discard practices, and reuse of cooking or campfire sites, may lead to recrystallisation. Thus, archaeologists must thoroughly examine and test all potential material for signs of recrystallisation prior to undertaking further analysis.

While modern comparative analogues can assist in evaluating the isotopic integrity of archaeological material (Kennett and Voorhies 1996), particularly in cases where contact with meteoric water is suspected, further steps must be taken in order to ameliorate results from latent inaccuracies associated with post-depositional change. A number of issues can be countered through careful selection of material using criteria suggested by Burchell et al. (2013b:262) – 1) a portion of the ventral margin must be intact and preserved; 2) the upper shell layer must be intact and preserved; 3) absence of evidence indicating exposure to fire, e.g. burnt portions of shell or association within a hearth feature; and 4) size of the fragment is sufficient to provide an adequate sample of growth. However, further measures must be taken in order to ensure specimens are not affected by recrystallisation. Thus, it has become common practice for analysts to subject material to X-ray diffraction (XRD), fourier transformation infrared spectroscopy (FTIR), cathodoluminescence or element chemical analyses, raman spectroscopy or Feigel's solution to determine the crystalline structure (aragonite or calcite) and assess the potential of recrystallisation prior to undertaking further analysis (e.g. Balmain et al. 1999; Burchell et al. 2013b; Fenger 2006; Friedman 1959; Goewert and Surge 2008; Hawkes et al. 1996; Kennett and Voorhies 1996; Petchey et al. 2013).

4.3 High-Resolution Sampling

An integral, yet often overlooked, step in stable isotope analysis is the extraction of calcium carbonates from target material. This process not only provides carbonate samples required for isotopic analysis but also plays a significant role in dictating the resolution and accuracy of resulting data. In molluscan shell, macro- and micro-growth structures provide a physical framework which facilitates the study of discrete time-scales ranging in length from sub-daily to annual (Hallmann et al. 2013). However, access to this high-resolution archive is limited by the techniques and technology employed to gather and analyse carbonate samples.

Inherent limitations can lead to decreases in attainable resolution and generation of ambiguous results. Among these, the notion of time-averaging is the most prevalent issue faced by researchers attempting to define discrete isotopic signals from accreting skeletal tissues (Goodwin et al. 2004).

Time-averaging refers to the homogenisation or dampening of isotopic signals owing to differences in the period of time represented by individual samples. Isotopic analysis of calcium carbonates using mass spectrometry requires powder samples to attain a minimum weight, dependent on the instrument being used, in order to produce an accurate signal (Merritt and Hayes 1994). As growth features in molluscan shell are often tightly spaced, especially micro-growth structures, researchers often find it necessary to utilise samples that represent more than a single point in time, sometimes stretching over multiple growth features. Eerkens et al. (2013) estimated that due to variability in growth rates a majority of their carbonate samples represented anywhere between one and four months of growth. Variations during this period are lost as delta values only represent an average of isotopic composition. Loss of extreme signals through smoothing is especially pertinent to studies modelling palaeotemperatures, where $\delta^{18}O$ values are used as a conversion variable to estimate temperature (see Grossman and Ku 1986). As a 1.0‰ change in $\delta^{18}O$ reliably equates to a 4-5°C variation in temperature (Grossman and Ku 1986), time-averaging can significantly alter results.

Changes in the rate of shell precipitation stemming from ontogenetic age can also lead to large differences in represented time, for instance samples from ontogenetically older sections of shell frequently correspond to larger time-slices due to slower precipitation causing the compaction of growth features (Burchell et al. 2013a; Burchell et al. 2013b). Size of the individual can produce similar patterns of shell formation, with larger mollusc species often exhibiting wider growth features and condensed lines and increments being common in smaller species. Consequently, appropriate sampling regimes must be selected in order to attain adequate resolution and ameliorate data sets from latent inaccuracies.

A review of the literature reveals a pattern of inadequate or inappropriate sampling techniques, which has potentially perpetuated issues associated with time-averaging. Specifically, studies that collect samples at arbitrary distributions (usually 0.5 – 2mm) are of the greatest concern (e.g. Aguirre et al. 1998; Bailey et al. 1983; Brockwell et al. 2013; Culleton et al. 2009; Kennett and Voorhies 1995; Kennett and Voorhies 1996). Arbitrarily spaced sampling can facilitate the latent mixing of seasonally derived growth features within individual samples, potentially introducing inaccuracies which may significantly alter interpretations. This issue is frequently exacerbated by the application of unsuitable sample extraction procedures.

The studies cited above utilise micro-drilling, a process in which a dentist's drill, or similar, is used to bore precise vertical holes at predetermined locations across the exterior or cross-section of a shell. The precision of these samples is constrained by the size of the drill bit, making it difficult to sample tightly spaced growth features (Wurster et al. 1999). Moreover, as carbonates are often sampled at intervals, arbitrary or otherwise, associated isotopic profiles are only representative of a series of snapshots taken through ontogeny with no information regarding the intervening time. Numerous studies have conducted comparisons between micro-drilling and other comparable, although potentially higher-resolution, carbonate extraction methods. In particular, Spötl and Mattey (2006) demonstrated that while both high- and low-resolution techniques can provide insight into environmental oscillations, micro-drilling failed to characterise several features and smoothed peak delta values by up to 1.5‰. This is not to suggest that micro-drilling should be dismissed out of hand, particularly as it has been applied effectively as part of recent archaeological studies (e.g. Burchell et al. 2013a; Burchell et al. 2013b), rather it should be seen as an example of the considerations that must be made when undertaking stable isotope analysis. Yet, few studies actively address or acknowledge the potential issues associated with sampling, meaning broad inaccuracies may be present throughout the literature.

Recent technological and methodological advances have permitted researchers to mitigate some of the issues stemming from time-averaging through high-resolution data recovery. Micro-milling allows greater spatial and temporal control to be maintained over sampling (Wurster et al. 1999). High temporal resolution is achieved by sampling from a series of transects placed parallel to growth structures using a computer-assisted rig (Figure 5) (Andrus and Rich 2008; Hinton 2012; Spötl and Mattey 2006; Wurster et al. 1999). This allows for an uninterrupted record of the shell's growth history to be obtained while simultaneously minimising the effects of time-averaging (Burchell et al. 2013b; Goodwin et al. 2004). Researchers have recently begun applying micro-milling in conjunction with micro-growth feature analysis to great effect, generating high-resolution reconstructions of palaeoenvironments (Carré et al. 2005a; Goodwin et al. 2001; Goodwin et al. 2003) and determinations of seasonal site use (Burchell et al. 2013a; Burchell et al. 2013b; Burchell et al. 2013c; Hallmann et al. 2013; Hallmann et al. 2009). It should be noted that micro-milling is not entirely immune to the effects of time-averaging with resolution still constrained by the size of the drill bit, although to a lesser extent than micro-drilling. Further, minimum sample weights required for mass spectrometry frequently leads to material from multiple adjacent transects being combined to produce a viable sample (e.g. Hanson et al. 2010). The destructive nature of powder sampling, both micro-milling and -drilling, is also problematic as it prevents multiple analyses from being undertaken on individuals and, in the case of archaeological material, damages irreplaceable specimens.

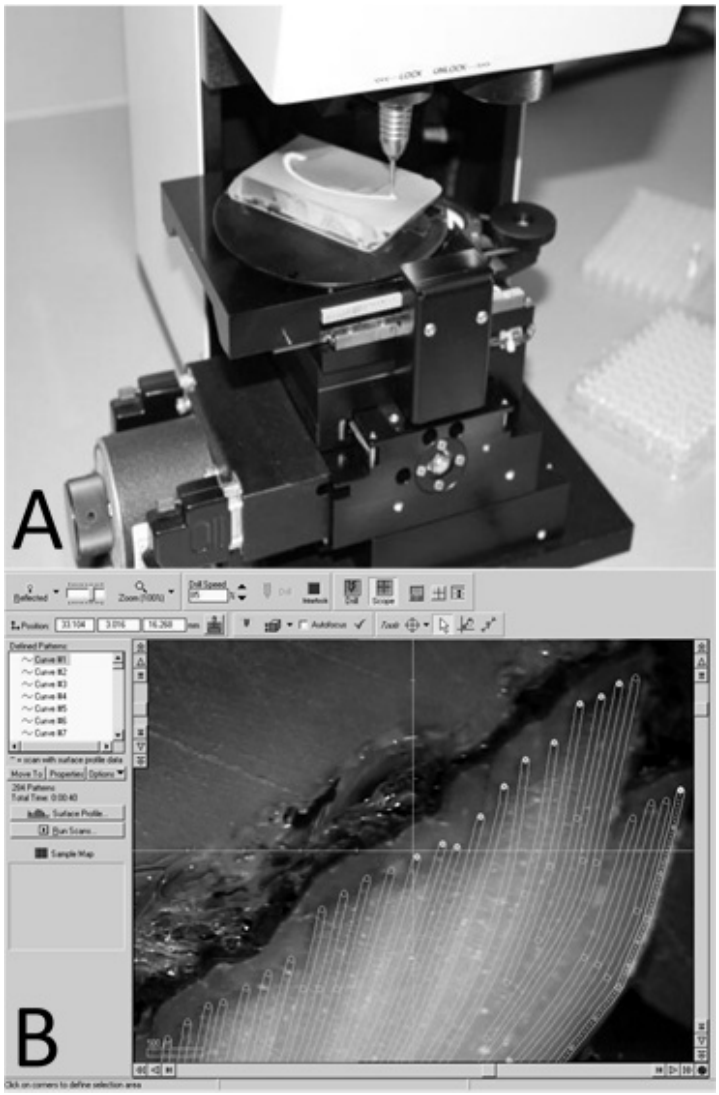

Figure 5 A) Computer-assisted NewWave MicroMill system. B) Digitised scan lines in preparation for micro-milling (images adapted from Hinton 2012:53-54).

To further counter issues associated with time-averaging as well as the destructive nature of carbonate powder collection, the efficacy of applying secondary ion mass spectrometry (SIMS), another high-resolution geochemical analysis technique, to stable isotope studies has been explored (e.g. Hanson et al. 2010; Weidel et al. 2007). SIMS works on the principle that charged particles (secondary ions) are ejected from a sample surface when bombarded by a beam of heavy particles (Griffiths 2008). This occurs in vacuum allowing the secondary ions to be channelled into a mass spectrometer for analysis. The size and intensity of the beam can be adjusted allowing for very precise sampling that causes very little damage to the specimen, spot sizes are often 10-15 μm in diameter and 2-3 μm in depth (Hanson et al. 2010; Weidel et al. 2007). Comparisons between SIMS and micro-milling used in conjunction with continuous flow isotope ratio mass spectrometry (CF-IRMS) have revealed that while micro-milling/CF-IRMS provide better precision, SIMS allows for greater temporal resolution (Hanson et al. 2010). While SIMS holds great potential for the generation of exceptionally detailed isotope profiles, for a majority of researchers micro-mills/drills remain more accessible and, provided methods have been properly tailored to approaching research questions, are more than capable of producing data at an appropriate resolution.

Comparisons between Stephens et al. (2008) and Hinton (2012) illustrate the impact different sampling methods can have on dataset accuracy. Stephens et al. (2008) utilises laser ablation which employs a focused laser to liberate CO_2 from carbonates either in a vacuum (or helium), the gas is then led directly into a mass spectrometer allowing rapid *in situ* analysis of material (Smalley et al. 1989). However, laser ablation does not provide the resolution attainable by micro-milling or even micro-drilling (see Spötl and Mattey 2006 for a comparison of the techniques). Heat produced by the laser creates a thermal halo around each ablation point limiting how tightly samples can be spaced and potentially altering the chemical composition of adjacent carbonates (Spötl and Mattey 2006; Stephens et al. 2008). This not only decreases precision and attainable resolution but also the amount of shell that can be analysed. Stephens et al. (2008) was forced to truncate sampling as growth features became too closely spaced to ensure individual growth structures were sampled. Conversely, Hinton (2012) employs micro-milling, enabling a continuous profile of modern and archaeological material to be generated. While this higher resolution data depicted a seasonally derived 'saw-tooth' pattern comparable to that found in Stephen et al's material, previously absent extreme amplitude changes were characterised. However, it is important to reiterate is that no sampling strategy is objectively better (or worse) than any other, rather that methodologies selected to complement research questions will provide more accurate and meaningful interpretations.

5. Future Directions for Archaeological Sclerochronology

Sclerochronological analyses provide unique opportunities for archaeologists to recover data at a resolution that is unattainable using more traditional methods. While the application of these techniques to archaeological contexts has seen increasing interest over recent years, basic limitations appear throughout the literature. Although the fundamental methodologies driving sclerochronology are well established, their incorporation into archaeological investigations is still a relatively new concept. Consequently, with a few recent exceptions, research applications are often underdeveloped. Recent practical and methodological advances potentially afford substantial increases in the attainable resolution and accuracy of data collection and analysis, yet these practices are rarely deployed. Thus, this paper outlines a series of recommendations that aim to increase the accuracy and validity of interpretations reached through the application of sclerochronology to archaeological material:

1. *Establish high-resolution localised environmental records to contextualise archaeological data*

The importance of contextualising archaeological data against localised understandings of environmental conditions has been demonstrated by numerous studies documenting varying intra- and inter-regional environmental conditions and their impact on mollusc carbonate precipitation and fractionation (Bernstein 1990; Gillikin et al. 2005a; Hallmann et al. 2008; Henry and Cerrato 2007; Jones and Quitmyer 1996). Yet, many researchers continue to rely upon regionally-scaled environmental data or extrapolate local data across

geographic boundaries (e.g. Brockwell et al. 2013). This normative practice distorts any interpretation of the archaeological record as it assumes uniform conditions and responses across landscapes and populations that may be substantially different (Claassen 1991). Thus, an increasing number of publications suggest that in order to reach meaningful interpretations of human-environment interactions the archaeological record must be contextualised within a framework of local palaeoenvironmental conditions (e.g. Ulm 2013; White 2011).

If archaeologists are to approach the sophisticated understandings of past human-environment relationships possible through sclerochronology, the emphasis placed on establishing detailed understandings of local environmental conditions must be substantially increased. While the very nature of data generated through sclerochronology provides the foundations for localised environmental records, it is up to researchers to acknowledge its importance and incorporate such understandings into interpretative frameworks. Yet, few published studies devote adequate time to discussing modern analogues and their use in conjunction with archaeological material. Increased transparency of these processes would better illustrate the advantages of including localised environmental information as the basis for archaeological interpretation, allowing researchers to make increasingly informed decisions regarding methodological choices.

2. Incorporate appropriate sample sizes to reduce costs and ensure accuracy

While employing sclerochronology allows the generation of high-resolution datasets, the application of these techniques can be a time consuming, laborious and expensive process. Researchers often commit significant resources to analysing large quantities of growth features as well as extracting and processing hundreds if not thousands of powder samples per assemblage (e.g. Burchell 2013:18). While numerous aids have been developed to assist in the rapid assessment of growth features, including acetate peels (Pannella and Copeland 1968) and staining solutions (Schöne et al. 2005a), carbonate sampling and analysis remains costly. Thus, commissioning the analysis of large collections can prove to be prohibitively expensive, particularly for researchers whose institutions lack the necessary equipment or expertise required for in-house processing.

In order to minimise costs, researchers frequently undertake rigorous analysis on a limited number of specimens, particularly modern material where it is common for only a handful of individuals to be assessed (e.g. Brockwell et al. 2013; Jew et al. 2013; Stephens et al. 2008). Although high-resolution data is gathered from these specimens, it is unlikely that the results are representative of broader trajectories owing to widely documented variability in local environments and intra- and inter-population responses to environmental conditions (Bernstein 1990; Gillikin et al. 2005a; Hallmann et al. 2008; Henry and Cerrato 2007; Jones and Quitmyer 1996). As interpretations of isotopic signals recovered from archaeological material rely on modern contextual data, limited sample sizes of modern specimens can lead to the introduction of latent biases and inaccuracies to archaeological interpretations. Thus, robust modern analogues cannot be sacrificed to minimise costs.

This paper posits that the incorporation of strong contextual foundations, which draw upon broad modern collections, not only allow significantly more accurate and meaningful archaeological interpretations but potentially reduce time and budgetary strain. Archaeological material is often heavily analysed with some researchers proposing that a series of samples per specimen is required to provide an adequate proxy for seasonal variation (Andrus and Crowe 2000; Mannino et al. 2003). While this is true for studies attempting to approach detailed palaeoenvironmental reconstructions or generate master chronologies, it is not necessary to maintain this resolution to address many other research questions – for example season of capture determinations only require targeted analysis of the ventral margin and adjacent areas (Culleton et al. 2009; Kennett and Voorhies 1996). In light of this, more robust understandings of modern environments and the impacts they have on contemporary shellfish populations may, in some cases, allow archaeologists to reduce the number of samples required from archaeological specimens, thus minimising cost and allowing larger archaeological assemblages to be analysed.

3. Select powder sampling methods suitable to research aims

Powder sampling regimes play a significant role in dictating the accuracy and resolution of stable isotope results. While micro-milling affords the highest available resolution it is also the most expensive and time-consuming, requiring the extraction and analysis of large quantities of carbonate samples. Micro-drilling requires less processing and analysis time owing to significantly fewer samples extracted from each specimen (Spötl and Mattey 2006), while laser ablation allows *in situ* analysis of material (Stephens et al. 2008). However, in their current states these methods are unable to match the fine scale analysis of micro-milling (Spötl and Mattey 2006), often lead to latent inaccuracies through time-averaging (Eerkens et al. 2013), and can limit the extent to which specimens can be analysed (Stephens et al. 2008). While newer methods, such as SIMS, produce exceptionally high-resolution isotope profiles and are significantly less destructive to the specimen, the equipment and expertise required is not yet widely available (Hanson et al. 2010). Thus, until methods exhibiting increased expediency and attainable resolution are accessible, a balance must be struck between cost and the accuracy or resolution required to address designated research questions.

A rarely considered solution to this involves deploying a combination of sampling regimes. Burchell et al. (2013a; 2013b; 2013c) applied both micro-milling and micro-drilling to analyse *Saxidomus gigantea* specimens. Micro-milling was undertaken at the ventral margin and adjacent areas where growth was found to have slowed significantly, while micro-drilling is utilised in shell portions exhibiting faster growth. This sampling regime was altered on a specimen-by-specimen basis to account for varying size and ontogenetic age. By employing this joint methodology Burchell et al. (2013a; 2013b; 2013c) were able to generate the high-resolution data required for accurate seasonality determinations, while minimising the cost of sampling extensive archaeological assemblages over multiple annual cycles. However, it must be noted that the validity of this methodology may not extend to other studies as it is highly tuned to the morphology of the target taxa. Previous studies provided extensive knowledge of local environmental cycles and their effects on constituent *S. gigantea* populations (see Gillikin et al. 2005a; Hallmann et al. 2009). Furthermore, the research undertaken by Burchell et al. focuses on determining the season of capture of archaeological shellfish, meaning precise isotopic analysis was only required at the ventral margin and adjacent areas. Research attempting more extensive palaeoenvironmental reconstruction would require a higher resolution sampling regime throughout ontogeny. Nevertheless, these studies demonstrate how methodologies can be tuned to minimise time and financial costs while still meeting research parameters and generating meaningful interpretations.

4. Utilise datasets generated by complementary techniques to contextualise ambiguous results

While sclerochronology provides information essential to advancing fundamental understandings of past environments and their impacts on human behaviours, these techniques sometimes require supplemental datasets to further contextualise results. For example, comparative data would assist in alleviating difficulties associated with oxygen isotope signals in regions subject to the dual influences of temperature and water constitution (see Andrus and Thompson 2012; Culleton et al. 2009), as well as those stemming from palaeotemperature equations requiring precise knowledge of $\delta^{18}O_{water}$ (see Epstein et al. 1953; Grossman and Ku 1986; McCrea 1950). Incorporating complementary techniques, such as trace element analysis and clumped isotope thermometry, may significantly increase the accuracy of palaeoenvironmental determinations. It must be noted however, that these techniques require further development before they can provide meaningful datasets.

6. Conclusions

The application of high-resolution data recovery techniques has become a fundamental component of broader archaeological frameworks focused on the investigation of human-environment relationships. Palaeoenvironmental proxies derived from archaeological deposits provide information relating to localised conditions and act as a direct link between environment and human behaviour. The fine scale datasets

derived from these contexts afford the opportunity for archaeologists to approach more detailed understandings of environmental conditions within temporally and spatially constrained frameworks. This in-turn informs more nuanced interpretations of the interactions between the environment, culture and human decision-making processes, allowing higher-order questions to be addressed. As a result, techniques such as sclerochronology are increasingly deployed in an attempt to increase archaeological resolution and approach better understandings. However, as with all novel methodologies it is essential to understand and acknowledge their complexities and limitations along with their advantages. In doing so researchers are able to unlock the full potential of these techniques while ameliorating results from a myriad of latent inaccuracies and biases.

Yet, the literature reveals that these challenges are often not met with the degree of thoughtfulness required to adequately address them. Instead they are often dismissed or ignored altogether. The effects of inappropriate methodology can be seen through the loss of important data and the introduction of a variety of uncertainties relating to the accuracy and validity of results. However, these challenges should not dissuade the use of sclerochronology, it should instead encourage researchers to take care in building methodological frameworks and understand the implications of those decisions. Provided appropriate precautions are taken sclerochronology offers a unique opportunity, enabling archaeologists to gain understandings of fundamental controls that assisted in structuring the everyday lives of past human populations and characterise the impacts of these decision-making processes.

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