MIDDLE HOLOCENE PERIODICITIES IN RAINFALL INFERRED FROM OXYGEN AND CARBON ISOTOPIC FLUCTUATIONS IN PREHISTORIC TROPICAL ESTUARINE MOLLUSC SHELLS*

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Stable oxygen and carbon isotopic ratios in modern and archaeological estuarine mollusc shells, Polymesoda radiata, change in accordance with seasonal salinity fluctuations in the Acapetahua estuary located on the Pacific coast of southern Mexico. This region receives ~ 3000 mm of precipitation annually, most during a wet season between April and October. The changing flux of fresh water and organic detritus into the estuary causes large changes in the oxygen and carbon isotopic composition of the estuarine waters and in the carbonate precipitated by P. radiata. Oxygen isotopic ratios in the shells of molluscs collected by late Archaic period populations (5000–4000 BP) in this region indicate that patterns of rainfall were similar to today. Modern shells, however, exhibit much more negative carbon isotopic values than observed in prehistoric shells. This change may be associated with the input of modern fertilizers into the estuary.

KEYWORDS: MESOAMERICA, ARCHAIC PERIOD, OXYGEN ISOTOPIC ANALYSIS, CARBON ISOTOPIC ANALYSIS, MOLLUSC SHELLS, POLYMESODA RADIATA

INTRODUCTION

This paper reports palaeoclimatic information based on stable oxygen and carbon isotopic analysis of estuarine mollusc shells found in late Archaic period (5000–4000 BP) archaeological deposits located on the Pacific coast of south-western Mexico (Fig. 1). These shell deposits represent some of the earliest evidence for coastal occupation in Mesoamerica, accumulating during a time when human populations were making the economic transition from hunting and gathering to agriculture. The primary goal of this study was to determine the environmental context of this important economic transformation.

The oldest-known sites in this area consist of six prominent shell mounds situated within the coastal wetlands. One of these, Cerro de las Conchas, has been dated to c. 6000 BP. This mound is presently located at the inland margin of a large (30–60 sq km; Helbig 1976, 219), freshwater swamp (El Hueyate or Cantileña); however, associated mollusca are either estuarine or marine, indicating large palaeoenvironmental changes since its formation. This site is being investigated by others (Clark 1994) and is not discussed here.

Our study focuses on material from the site of Tlacuachero, one of five shell mound sites that constitute sizeable islands in the Acapetahua estuary immediately to the north-west
Figure 1  Study area (southern Mexico) showing late Archaic period shell mound sites, coastal lagoons of the Acapetahua estuary, the El Hueyate swamp and coastal rivers discussed in this paper.
of the El Hueyate swamp (Fig. 1). These late Archaic period deposits, including Tlacuachero, accumulated between 5000 and 4000 BP, slightly later than the Cerro de las Conchas shell mound. Because the archaeological constituents of all six shell mounds are similar, they are believed to have been formed by the same prehistoric population that we call the Chantuto people. Subtle differences in the tool kits differentiate early from late Chantuto cultures (Blake et al. 1991).

The modern Acapetahua estuary consists of five interconnected coastal lagoons surrounded by a mangrove forest. The mangroves reach 25m in height (Helbig 1976, 217), making this mangrove formation one of the most developed on the Mexican coast. Cattail marshes occur at the inland margins of the mangrove swamp. The lagoons are shallow (~2 m), have soft bottoms and are brackish (salinity range ~2 to 24%). The lagoonal waters are affected both by tides and the input of fresh water from rivers that empty into their heads.

The lagoons of the Acapetahua estuary (sometimes called Laguna del Viejo: e.g., Contreras 1985, 117) are the southernmost of approximately 63 coastal lagoons occurring along the west coast of continental Mexico from the Rio Colorado to the Guatemala border (Contreras 1985). These lagoons were formed during the Holocene marine transgression. Our findings concerning the history of the Acapetahua estuary, therefore, have wider significance for the Pacific coastal lagoons of Mexico.

The wetlands in this region are bounded on the seaward side by a three-kilometre-wide band of relict shoreline reflecting late Holocene progradation of the coastline. These relic shorelines have not yet been dated precisely either by archaeological (e.g., Gagliano 1984, 33) or radiometric approaches. Progradation of the coastline has protected the archaeological sites in the wetlands that otherwise would have been destroyed by coastal processes (e.g., Waters 1992, 272).

The biotic constituents of the archaeological sites in the present-day Acapetahua estuary indicate the presence of a nearby estuarine environment during their formation in the late Archaic period. The deposits are dominated by marsh clam (Polymesoda radiata; Severeyn 1993) shells and lack a soil matrix. Small quantities of other molluscs have been found, all of which co-occur with marsh clams, and were probably inadvertently carried by people to the sites with the clams. Similarly, most of the vertebrate material identified in the archaeological deposits of the late Archaic period are from animals found today in the wetlands. A few animals might have been taken offshore, or more likely, scavenged from the beach (Voorhies 1976; Anikouchine 1990). Finally, all identified phytoliths from the site of Tlacuachero (palm, sedge, grass, Heliconia sp. and possibly bamboo; Jones 1988) are found in the estuary today or close by. Accordingly, the existing archaeological data suggest palaeoenvironmental conditions during formation similar to those of today, but this requires more quantitative verification.

OXYGEN AND CARBON ISOTOPES AS PALAEOENVIRONMENTAL INDICATORS

Oxygen and carbon isotopic analysis of mollusc shells is a well-established approach for palaeoenvironmental reconstruction. In marine environments oxygen isotopic fluctuations have been used successfully as a palaeothermometer (Epstein et al. 1953). Shackleton (1973) analysed the oxygen isotopic composition of the sequential growth increments of Patella tabularis, a limpet found in the intertidal zone along the west coast of South Africa. He
D. Kennett and B. Voorhies determined that the amplitude of oxygen isotopic change paralleled annual variations in water temperature. Killingley and Berger (1979) were able to show close correlation between oxygen isotopic variations in modern mussel (*Mytilus californianus*) shell with variations in sea surface temperature of southern California.

While fluctuations in ocean temperature cause predictable changes in the oxygen isotopic composition of marine mollusc shells, predicting changes in estuarine mollusc shells is more complicated. This is because major changes in the oxygen isotopic composition of estuarine water can be caused by the influx of river water, as well as by seasonal temperature changes. The influx of river water affects the oxygen isotopic signal of estuarine water because continental water has lower concentrations of the heavier oxygen isotope ($^{18}O$) than ocean water (Keith *et al.* 1964). The complex interaction of these factors often makes it difficult to decipher the environmental causes of oxygen isotopic signals recorded in the shells of estuarine molluscs, particularly in temperate areas. For this reason, Deith (1983, 69) argued that oxygen isotopic analysis is useful as an environmental indicator only for open ocean molluscs and the analysis is unsuitable for estuarine species. However, in some estuarine settings seasonal variations in the isotopic composition of water, caused by the discharge of fresh river water, overwhelm the temperature effect (Dettman and Lohman 1993). In these settings, oxygen isotopic analysis of molluscan shells is potentially a valuable tool for reconstructing ancient water regimes.

Carbon isotopic changes ($\delta^{13}C$) in molluscs are more difficult to interpret and for this reason have received less study than oxygen isotopes (Wefer and Killingley 1980). Unlike oxygen isotopes, carbon isotopic variations are not temperature dependent. Carbon isotopic ($\delta^{13}C$) changes in the shells of marine molluscs have been found to reflect a combination of two primary factors: (1) the carbon isotopic composition of the water, and (2) the metabolic activity of the mollusc (Revelle and Fairbridge 1957). Killingley and Berger (1979) recognized that carbon isotopic concentrations in California mussel shells generally correlate with changes in the marine carbon reservoir caused by coastal upwelling. Although these carbon isotopic fluctuations were associated with seasonal changes in the isotopic composition of the water through changes in upwelling, the signal in the shell was slightly out of phase. This difference was explained as a metabolic effect.

In estuaries and marine lagoons the $\delta^{13}C$ composition of the water can also change according to relative concentrations of fresh and salt water. This is because differences can occur between the marine and terrestrial carbon reservoirs. On average, the $^{13}C/^{12}C$ isotopic ratio of ocean water is $-2\%$, whereas carbon isotopic ratios in fresh water, although highly variable, are generally much more negative ($<-8\%$) than marine concentrations (Keith *et al.* 1964). Therefore, freshwater influx into marine lagoons will affect $\delta^{13}C$ values.

On average, this region of the Pacific coast of south-western Mexico receives 3200 mm of rainfall annually (based on measurements taken by the Comisión Nacional de Agua at Escuintla between 1975 and 1990). Most of this rain falls between the months of April and October, with negligible amounts falling during the rest of the year. During the rainy season the rivers that transect the Soconusco coastal plain swell and flood the coastal estuaries with fresh water. Because fresh water is isotopically ($\delta^{18}O$ and $\delta^{13}C$) more negative than open ocean water, it should alter the isotopic composition of the Acapetahua estuary in a predictable way. Being in the tropics, seasonal fluctuations in offshore water temperatures
are small (~3 °C) and contribute less to variations in the oxygen isotopic signal (Hastenrath and Lamb 1977). We hypothesize that during wet season months estuarine waters should have higher concentrations of the lighter $^{16}$O and $^{12}$C isotopes, compared with the dry season. This seasonal variability should be recorded in the shells of the marsh clam Polymesoda radiata.

**MATERIALS AND METHODS**

*Modern clam shells*

Living specimens of *P. radiata* (Fig. 2) were collected from the Acapetahua estuary to study relations between fluctuating environmental factors and the oxygen and carbon isotopic variations during growth of these bivalves. Ten monthly collections of *P. radiata* were taken from the Los Cerritos lagoon (Fig. 1) during one annual cycle spanning 1989 and 1990. Water samples were collected from the lagoon at the same time. The $\delta^{18}$O of the water samples was determined by Michael DeNiro, Department of Geological Sciences, University of California, Santa Barbara, and are expressed relative to standard mean ocean water (SMOW). The salinity of these monthly water collections was determined using a field refractometer.

Five right valves of *P. radiata* were randomly selected for analysis from each of the monthly collections. The shells were thoroughly cleaned to remove any extraneous organic material that might contaminate the samples. All obvious organic material present on the surface of the shell, including the periostracum, was removed using a clean razor blade. The shells were rinsed in deionized water and dried at 85 °C. This process was repeated until all visible organic material was removed.

*Archaeological clam shells*

The archaeological specimens of *P. radiata* analysed came from the site of Tlacuachero (CAP-7; Fig. 1). In the field we collected 10 cm$^3$ of sediment, including shells, at 10 cm intervals within a vertical column. The whole clam valves used in this study were taken from six of these sediment samples spaced approximately one metre apart (Fig. 3).

Two archaeological clam shells were selected from each level for oxygen and carbon
isotopic analysis. Each valve was rinsed with deionized water to remove the adhering soil matrix and oven dried at 85 °C.

The shell margin of each modern and archaeological specimen, representing the final stages of growth prior to harvesting, was removed with a clean razor blade. To reconstruct periodicities in the estuarine environment throughout the life of these marsh clams we analysed subsurface samples of carbonate extracted along the growth line of several archaeological ($N = 12$) and modern shells ($N = 6$). Carbonate samples were extracted in 1.5 mm increments along a line from the shell margin to its hinge. These samples were used to reconstruct the oxygen and carbon isotopic variation during the life of each mollusc. A 0.5 mm dental drill was used to extract the samples.

Each sample of shell was ground into a fine powder using a mortar and pestle, loaded into a small copper ‘boat’ and roasted, under vacuum, at 400 °C for one hour. This procedure oxidises any remaining organic material. After roasting, the calcium carbonate
Middle Holocene periodicities in rainfall

was reacted in orthophosphoric acid at 90 °C, which was dried under vacuum at 100 °C for three days prior to the analyses. Oxygen and carbon isotopic ratios of the resulting CO₂ were determined using mass spectrometry (Finnegan MAT-251-mass spectrometer, Department of Geological Sciences at the University of California, Santa Barbara). Measurements are expressed in δ (delta) notation where:

\[ \delta^{18}O_{\%o} = \left( \frac{\delta^{18}O_{\text{sample}}}{\delta^{18}O_{\text{standard}}} - 1 \right) \times 1000 \]

\[ \delta^{13}C_{\%o} = \left( \frac{\delta^{13}C_{\text{sample}}}{\delta^{13}C_{\text{standard}}} - 1 \right) \times 1000 \]

All measurements are calibrated to PDB, the internationally accepted standard. More negative δ values indicate higher proportions of the 'lighter' ¹⁸O and ¹²C isotopes compared to the 'heavier' ¹⁸O and ¹³C isotopes, and vice versa. The precision of the oxygen and carbon isotopic measurements is better than 0.1‰.

Diagenesis

In order for this method to be reliable we needed to establish that there had been no postdepositional contamination of the shell carbonate. Although the shells from Tlacuachero appeared to be well preserved, we performed several procedures to screen archaeological specimens for diagenesis. Thin-sections of modern and archaeological shells were first examined with conventional and scanning electron microscopes. Visual inspection of the crystalline structure of the archaeological shells indicated that they were well preserved. Modern and archaeological shells also were examined using X-ray diffraction. The crystal lattice of contemporary *Polymesoda radiata* shells consists of aragonite. X-ray diffraction determined that the primary aragonite in the archaeological specimens was intact and that secondary deposits of calcite, indicative of diagenesis, were absent.

The outer surfaces of fossil shells are particularly subject to recrystallization. This process is generally associated with chemical exchange with percolating ground water (Shackleton 1973). Bailey *et al.* (1983, 397) recommended etching the outer surface of each archaeological specimen with a dilute acid to remove any diagenetic material. We performed an oxygen isotopic profile on an archaeological specimen before and after etching its surface with a dilute solution of HCl. The results, with and without the treatment, were the same (within 0.1‰) and indicate that the exterior surface of this archaeological shell is well preserved. However, shells from the same deposit can be subject to different rates of diagenesis, so each shell was cleaned using a dilute acid.

RESULTS

The upper line in Figure 4 (a) summarizes the range of δ¹⁸O variation of the monthly modern shell margin samples from September 1989 to August 1990. This represents an annual δ¹⁸O range of 4‰ (≈ −9‰ between the months of July and January and ≈ −5‰ between the months of February and June). The annual range of oxygen isotopic composition of shell margin carbonate is greater (4‰) than that of any single month (1‰). The δ¹⁸Osub PDB of the shell margin carbonate parallels the δ¹⁸Osub SMOW of the water...
During 1989-1990, the δ¹⁸O composition of modern shell margins (N = 5/month) plotted against: (a) the δ¹⁸O of estuarine water samples collected from the surface of the Los Cerritos lagoon at the same time; (b) the salinity of estuarine water samples taken from the surface of Los Cerritos lagoon; (c) average monthly rainfall measured on the coastal plain (Escuintla, Comisión Nacional de Agua) between September 1989 and August 1990; (d) water flow in the Cintalapa River (Comisión Nacional de Agua) between September 1989 and August 1990.

Samples collected each month from the Los Cerritos lagoon (Fig. 4 (a)). Oxygen isotopic composition of the estuarine water is higher (more positive) between January and June (~ -3 to -5) and lower from July to December (~ -6.1 to -7.8‰).

Successive changes in the oxygen isotopic composition of both shells and water are inversely correlated with salinity fluctuations in the Acapetahua estuary (Fig. 4 (b)). The oxygen isotopic values of the estuarine water and the shell margin carbonate are more positive (~ -4.2 to -6.2) when the water is more saline (18–20‰) between February and June (dry season). Oxygen isotopic values are consistently more negative (~ -7 to -9.2) between July and January when the estuarine waters are virtually fresh (1–2‰).

Figure 4 (c) compares the oxygen isotopic composition of shell margin carbonate and rainfall for the same time period. The average rainfall gauged at the Escuintla meteorological station on the coastal plain between September 1989 and August 1990 was 3370 mm and ranged from 865 mm in September of 1989 to 4.8 mm in January of 1990. There is a general relationship between greater amounts of the lighter ¹⁶O isotope (~ -9‰) and the wet season.
Middle Holocene periodicities in rainfall

Figure 5  Oxygen and carbon isotopic profiles of a modern clam shell collected from the Los Cerritos lagoon in August 1990.

(March–October). However, there is a lag of approximately three months before the signal is registered in the shell carbonate. This lag also can be seen when the oxygen isotopic composition of the shell carbonate is compared to the velocity of the Cintalapa River flowing into the Los Cerritos lagoon (Fig. 4 (d)). During this particular annual cycle (1989–1990) water velocity was highest (55 m$^3$/sec) in the month of September 1989. This correlates with more negative oxygen isotopic ratios (inferred less saline water) in the shell margin samples of *P. radiata* and the estuarine water. Water flow decreased rapidly after September and remained low (2–4 cm$^3$/sec) until March of 1991 when it began to increase again.

Oxygen isotopic variation in the growth increments of all shells analysed record successive fluctuations in estuarine salinity. The amplitude of oxygen isotopic change in modern shell profiles is consistent with the range exhibited in the monthly shell margin samples. An oxygen isotopic profile for a modern marsh clam collected in August of 1990 is displayed in Figure 5. The amplitude of oxygen isotopic change in the two years of growth represented by this profile is 6‰, ranging from −3.5 to −9.5‰. The oxygen isotopic value at the shell growth margin is −7.01‰, which is consistent with other specimens of *P. radiata* collected from the Los Cerritos lagoon in August of 1990 (see Fig. 4 (a)).

Carbon isotopic values in all shells examined also appear to oscillate in consonance with seasonal changes in estuarine salinity. This is suggested by carbon isotopic values paralleling fluctuations in oxygen isotopes. The carbon isotopic profile displayed in Figure 5 exhibits a $\delta^{13}C$ range of ~3‰ from −7.865 to −10.910‰ and is positively correlated with the oxygen isotope profile. This pattern is also clear in all of the archaeological shells examined (e.g., Fig. 6). Carbon isotope composition of the shell growth margin (−10.910‰) is similar to that of other shells collected during this month.
Figure 6  Oxygen and carbon isotopic profiles of an archaeological clam shell from the late Archaic period shell deposits at Tlacuachero (level 1.20–1.40 m).

The oxygen isotopic profiles of all analysed archaeological and modern shells are similar (Fig. 7). Marsh clam shells from the late Archaic period deposits exhibit mean oxygen isotopic values between $-7\%o$ and $-8\%o$, whereas the mean oxygen isotopic value of all modern shells was $-7\%o$. In general, the amplitude of annual oxygen isotopic variation in

Figure 7  Summary of the range and mean of the $\delta^{18}O$ measurements from analysed archaeological shells from Tlacuachero ($N = 2$ per level) and modern ($N = 6$) specimens of Polymesoda radiata from the Los Cerritos lagoon. For each excavation level investigated, a line shows the range and a dot indicates the mean of the values for profiles of two individual clams. In addition, the mean and range of the six modern shell profiles are shown.
the archaeological shells is comparable to the modern forms. Archaeological *Polymesoda radiata* shells collected from the uppermost, Archaic period deposits (1.20–1.40 m; c. 4000 BP) at Tlacuachero deviate slightly from this pattern. Dry season conditions apparently were similar, but the more negative δ values during the wet season (2‰ more negative) are indicative of slightly greater rainfall during this time compared to today.

The δ¹³C ranges and means of all archaeological shell profiles from Tlacuachero are also similar (Fig. 8). However, there is a significant difference in the mean carbon isotopic value (δ¹³C) for the archaeological clam shells compared with the mean of the modern shells. The δ¹³C of the modern clams is approximately 4‰ more negative than the mean of all the archaeological shells studied (modern = −10.201‰; archaeological = −6.419‰).

**DISCUSSION AND CONCLUSIONS**

Our analysis of modern *P. radiata* show that change in the oxygen isotopic composition of shell carbonate accurately reflects fluctuations in estuarine salinity caused by seasonal changes in rainfall. Comparison of the oxygen isotopic (δ¹⁸O_{SMOW}) composition of estuarine water with the δ¹⁸O of shell margin samples indicates that the carbonate precipitated by the mollusc is in isotopic equilibrium with the water. Changes in the oxygen isotopic composition of the water and shell margin carbonate are clearly related to fluctuations in estuarine salinity. Estuarine salinity fluctuations are controlled by the intensity of rainfall on the coastal plain and bordering Sierra Madre mountains, although there is a lag of approximately three months before the changes in rainfall affect estuarine salinity. Presumably the lag between rainfall and estuarine salinity occurs because at the end of the rainy season the influx of fresh water continues after the monsoonal rains cease.

Prehistoric shells from the stratigraphic levels at Tlacuachero (CAP-7) exhibit oxygen isotopic profiles that are similar to modern specimens. This indicates that the pattern of
rainfall was much the same throughout the late Archaic period (5000 to 4000 BP) and also was similar to today's conditions. Michaels and Voorhies (forthcoming) argue that human groups in this region at this time were mainly hunter-gatherers, possibly practicing some form of incipient horticulture. The Chantuto middens have been interpreted as specialized activity locations periodically used for extracting shellfish, fish and possibly shrimp from the Acapetahua estuary. Like most hunting-gatherers, populations in this region during the late Archaic period must have been closely tied to the available natural resources of the terrestrial and littoral environments. The large fluctuations in the intensity and timing of rainfall would have caused significant changes in the distribution of resources. Humans would have responded to such changes. The bedded deposits at Tlacuachero and other littoral shell middens dating to this time interval appear to be relatively uniform and are thought to be indicative of a stable subsistence strategy (Michaels and Voorhies forthcoming). Stable patterns of middle Holocene rainfall in this region must have contributed to the stability of human subsistence and settlement strategies.

Significant changes in rainfall did not occur at the end of the late Archaic period, or rather, at the time that the uppermost samples that we have studied were deposited. However, it is clear that populations living on the Pacific coastal plain during the succeeding Preclassic period did change their subsistence strategies. Hunting and gathering was complemented by intensive maize agriculture and higher levels of socio-political complexity began to develop (e.g., Clark 1991). Our results suggest that this socio-economic transformation was not caused by environmental fluctuations at the end of the late Archaic period.

Carbon isotopic fluctuations in modern and archaeological shells are positively correlated with oxygen isotopic changes. Lower δ13C concentrations are associated with lower δ18O and, conversely, higher δ13C concentrations co-occur with higher δ18O values. This relationship suggests that changing carbon isotopic values are caused by the influx of isotopically lighter water into estuaries during wet season months. Changes in organic input (relatively light δ13C) into the estuaries related to changing fresh water influx also contribute to fluctuations in the isotopic signal.

Fluctuating metabolic rates in molluscs can also affect carbon isotopic (δ13C) fractionation. Wefer and Killingley (1980) reported a positive relationship between the δ18O and the δ13C in strombid snails from the coast of Bermuda. They suggested that this relationship is partly due to metabolic changes causing changes in shell growth rate. The lighter carbon values (more 12C) occurred during summer months when shell growth rates were high. This also appears to be the case with the marsh clam Polymesoda radiata. More negative δ13C values occur during wet season months when shell growth rates increase. Further research is required to determine if all variables contribute equally to the carbon isotopic signal or if one dominates the others. Regardless of the cause, we have found that higher concentrations of the lighter 12C isotope occur during wet season months and are positively correlated with the δ18O signal.

The δ13C profiles of prehistoric shells also indicate environmental stability through the late Archaic period. However, the average value of δ13C in archaeological shells is significantly lower than the modern shells analysed. We suggest that this difference in carbon is the result of more intensive habitation of the coastal plain, possibly related to the use of chemicals and fertilizers by the modern farmers in the region. Although further research is needed to determine why the stable carbon isotopic composition of modern shells is different, this method potentially could be used to monitor modern wetland environments.
Finally, although there are a variety of methods potentially available for seasonal determinations of prehistoric coastal sites, archaeologists working on shell middens in tropical settings have encountered difficulties because of the relative lack of seasonality. Identification of seasonal oxygen and carbon isotopic oscillations in molluscs, related to patterns of rainfall, provides a means for determining the season of prehistoric shellfish harvesting in tropical estuarine settings. Preliminary seasonality studies indicate that these molluscs were collected at all times during the year by the prehistoric people that occupied this location but that preferred seasons shifted over time.

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