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# **$^{18}\text{O}$ fractionation in barnacle calcite: a barnacle paleotemperature equation**

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## ABSTRACT

The calcitic shells of sessile barnacles (Balanomorpha) taken from ocean waters ranging in temperature from  $-2^{\circ}$  to  $26^{\circ}\text{C}$  have a near constant  $^{18}\text{O}$  enrichment compared with mollusk values assumed to be at thermodynamic equilibrium. The enrichment, about 1.3‰ as  $\delta^{18}\text{O}$ , will cause estimates of ancient ocean temperatures from  $^{18}\text{O}$  analyses on fossil barnacle shells to be about  $5^{\circ}\text{C}$  too cold unless a modified paleotemperature equation is used.

## 1. Introduction

Temperature-related variations in the  $^{18}\text{O}/^{16}\text{O}$  ratio of calcium carbonate precipitated from water were investigated in the pioneering work of Urey (1947) and Epstein *et al.* (1953). Epstein *et al.* proposed a paleotemperature equation based on isotopic fractionation behavior in mollusk calcite which is commonly accepted as describing thermodynamic equilibrium of the calcite-water system. Isotopic studies on various mollusks have demonstrated that the paleotemperature equation is applicable over a wide range of growth conditions (Lowenstam and Epstein, 1954; Emiliani and Mayeda, 1964; Longinelli *et al.*, 1962; Lloyd, 1967), although some exceptions have been observed (Tourtelot and Rye, 1969; Wefer and Killingley, 1980). This "mollusk" paleotemperature equation is widely applied in deep-sea stratigraphy and paleoceanography (Vincent and Berger, 1981) and such work has largely focused on the isotopic composition of foraminifera.

In most cases where disequilibrium isotopic fractionation has been observed, the measured  $^{18}\text{O}$  levels, expressed in the usual notation as  $\delta^{18}\text{O}$  values, have been lower than equilibrium values. Such fractionation has generally been attributed to "vital effects" (Urey *et al.*, 1951) involving kinetic fractionation and  $^{18}\text{O}$  depletion by various metabolic pathways. For example, extensive research on corals has shown that their aragonitic exoskeleton is generally depleted in  $^{18}\text{O}$  with respect to expected equilibrium values. Various reasons for such fractionation behavior have been proposed, including the effect of algal symbionts in hermatypic corals and the

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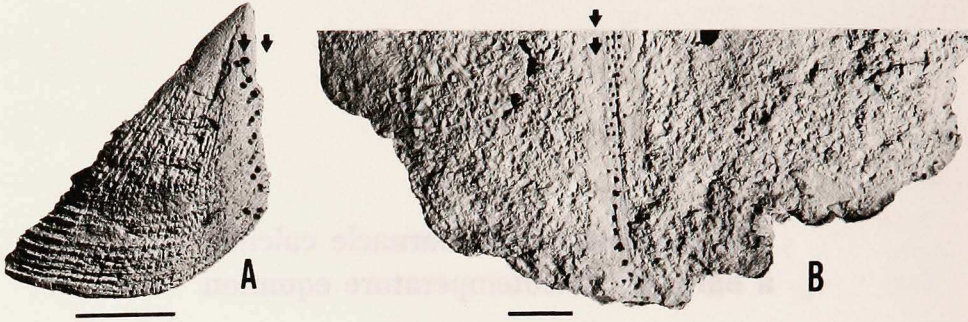


Figure 1. Exterior of right scutum (A) and surface of calcareous basis that was cemented to the substratum (B) of a large sessile barnacle, *Balanus aquila*, from Monterey, California. Samples for  $^{18}\text{O}/^{16}\text{O}$  determinations were taken from the oldest (top, arrows) to the youngest portions (bottom) of each shell (cf. Fig. 2). Scale bars equal 1 cm.

incorporation of  $^{18}\text{O}$  depleted metabolic  $\text{CO}_2$ . Although  $\delta^{18}\text{O}$  values for some corals have been shown to be systematically offset from equilibrium within genera, the "vital effects" may be different from one genus to another (Weber and Woodhead, 1970; Weber and Woodhead, 1972; Erez, 1978).

In contrast to extensive work on other calcareous organisms, very few isotopic measurements have been reported (Keith *et al.*, 1964; Killingley, 1980; Killingley *et al.*, 1980) on a large subclass of the Crustacea, the Cirripedia, or barnacles. Numerous barnacles co-occur with various mollusks in marine environments throughout the world, and their remains can constitute as much as 50% of the carbonate sediment in certain continental shelf areas (Milliman, 1972; Piessens and Lees, 1977). We show here that balanomorph barnacles secrete their exoskeletal calcite with a systematically higher  $^{18}\text{O}$  content than the corresponding mollusks over a wide range of temperatures.

## 2. Materials and methods

A suspicion that barnacle calcite might be enriched in  $^{18}\text{O}$  relative to mollusks arose in a study of both from the hydrothermal field at 21N off the west coast of Mexico (Killingley *et al.*, 1980). Therefore we chose a very large specimen (12 cm wide, 8 cm high) of the barnacle *Balanus aquila* (cf. Newman and Abbott, 1980) from the low intertidal at Monterey, California, collected June 30, 1961, and where daily records of seawater temperature and salinities were available. Closely spaced samples were taken with a 0.5 mm drill along a radius of the basis and also from the inner sheath and from one of the opercular plates of the same specimen (Fig. 1). They were isotopically analyzed by standard procedures with a precision of 0.1‰ (Berger and Killingley, 1977).

To establish whether the isotopic values found in the *B. aquila* specimen were

indicative of a general phenomenon, we analyzed 11 shells of sessile barnacles (10 genera, 11 species) from various locations around the world which included intertidal and subtidal forms (Table 1). Intertidal barnacle species only grow when immersed (Bourget and Crisp, 1975) and none of the specimens used in this study was from tide pools. Typically we measured isotopic profiles (Killingley, 1981) representing at least one year's temperature range, which could then be compared with the corresponding temperature minima and maxima in the field. We also measured isotopic profiles on two mollusk species co-occurring with two of the barnacles. Two species of pedunculate or stalked barnacles were also analyzed isotopically. X-ray diffraction analyses were carried out on samples of all specimens in the study to estimate the Mg content of the calcite. Although we experimented with various pre-treatments of the samples before isotopic analyses, including heating at 350°C under vacuum and soaking in  $\text{H}_2\text{O}_2$  solution, we found no sensible difference in results between these treatments and merely air-drying at 60°C. Therefore all data are on air-dried drill samples.

Equilibrium  $\delta^{18}\text{O}$  values for calcite were estimated using the paleotemperature equation (Epstein *et al.*, 1953); the temperature for the growth site; and the water isotopic composition obtained from salinity- $\delta^{18}\text{O}$  relationships (Craig and Gordon, 1965). Data sources for temperature and salinity are given in Table 1.

### 3. Results and discussion

The  $\delta^{18}\text{O}$  isotopic profile from the basis of the barnacle *B. aquila* is plotted in Figure 2. The values are remarkably high compared to calculated equilibrium values that are also shown in Figure 2. The  $\delta^{18}\text{O}$  results on the inner sheath and on an opercular plate from the same specimen are also much higher than expected and similar to the basis values (Fig. 2). The profiles shown for *B. aquila* represent approximately 60, 40, and 100% of the total sequential growth for the basis, sheath, and opercular plate, respectively. The rather flat profile for the plate is probably due to an averaging effect caused by low resolution sampling of a small piece of material. We did not attempt to match the time scale of amplitude variations with the calculated curve (because the growth rate probably decreased markedly with age, the 1961 maximum isotopic value may have been missed in our sampling). The main purpose was to compare the vertical range of amplitude variations.  $\delta^{18}\text{O}$  results on the other sessile barnacles that we analyzed also showed a clear trend toward heavy  $^{18}\text{O}$  values similar to *B. aquila*. Each isotopic profile showed a range of  $\delta^{18}\text{O}$  values that was generally consistent with the expected range (within analytical error) for the growth location but was offset to higher  $\delta^{18}\text{O}$  values by at least 1‰. (It is important to obtain the expected range of  $\delta^{18}\text{O}$  values to demonstrate year-round growth of the shells.) Maxima and minima results for these species are shown in Table 1.



Table 1.

Species, locality and depth	Water temperature °C	Salinity ‰	Water $\delta^{18}\text{O}$	Calculated equilibrium $\delta^{18}\text{O}$	Measured $\delta^{18}\text{O}$	$\Delta\delta$	Temperature and salinity data source*
<b>CIRRIPIEDIA (CRUSTACEA)</b>							
Lepadomorpha Pilsbry							
Scalpellloidea Zevina							
<i>Neolepas zeviniae</i> Newman Eastern Pacific, 20°50'N; 109E, 2600 m	1.8	34.6	-0.3	3.65	4.4	0.75	1
<i>Calantica villosa</i> (Leach) New Zealand, 45°30'S; 171E, intertidal	Max. 16.0 Min. 6.4	34.6 34.0	-0.2 -0.65	Min. -0.10 Max. 1.90	0.91 2.12	1.01 0.22	2
<i>Pollicipes polymerus</i> Sowerby California, 32°45'N; 117°20'W, intertidal	Max. 21.50 Min. 14.0	33.65 33.50	-0.60 -0.70	Min. -1.70 Max. -0.10	-0.98 0.62	0.72 0.72	3
Balanomorpha Pilsbry							
Chthamaloidea Darwin							
<i>Euraphia hembeli</i> Conrad Hawaii, 19°45'N; 155°05'W, intertidal	Max. 26 Min. 23.5	34.7 34.7	0 0	Min. -2.1 Max. -1.6	-0.95 -0.21	1.15 1.39	4
Coronuloidea Leach							
<i>Chelonibia testudinaria</i> (Linn) French Frigate Shoals, 23°50'N; 166°10'W, on turtle	Max. 26 Min. 23	35.0 35.0	0.1 0.1	Min. -1.95 Max. -1.45	-0.73 -0.10	1.22 1.35	4
<i>Bathylasma corolliforme</i> (Hoek) Ross Sea Shelf, 77°45'S; 166°25'E, 30 m	-1.8 ± 0.3			3.55	4.58	1.02	5
<i>Bathylasma hirsutum</i> (Hoek) N. Atlantic, 47°42'N; 08°14'W, 1,200 m	8.3	35.3	0.15	2.17	3.36	1.19	6
<i>Tetrachaelasma southwardi</i> Newman & Ross Seamount off Madagascar, 26°29'S; 46°07'E, 2,000 m	2.5	34.7	-0.2	3.55	4.85	1.30	7

<i>Epopella plicata</i> (Gray)	Max. 16.0	34.6	-0.2	Min. -0.10	1.32	1.42	2	
New Zealand, 45°30'S; 171E, intertidal	Min. 6.4	34.0	-0.65	Max. 1.90	3.01	1.11		
<i>Tetraclita rubescens</i> Darwin	Max. 21.50	33.65	-0.60	Min. -1.70	-0.24	1.45	3	
California, 32°45'N; 117°20'W, intertidal	Min. 14.0	33.50	-0.70	Max. -0.10	1.24	1.14		
Balanoidea Leach								
<i>Semibalanus cariosus</i> (Pallas)					0.46	1.75	1.29	4
Washington State, 48°25'W; 125W, intertidal								
<i>Chirona evermanni</i> (Pilsbry)	Max. 4.5	33.1	-0.9	Min. 2.22	3.42	1.20	4	
Bering Sea, 54°59'N; 165°44'W, 120 m	Min. 3.0	33.1	-0.9	Max. 2.65	3.92	1.27		
<i>Balanus aquila</i> (Pilsbry)	Max. 15.6	33.72	-0.50	Min. -0.30	1.05	1.25	8	
California, 35°50'N; 121°55'W, intertidal	Min. 11.9	33.89	-0.40	Max. 0.72	2.02	1.45		
<i>Austromegabalanus nigrescens</i> (Lam.)	Max. 24	35.5	0.3	Min. -1.35	-0.32	1.03	9	
W. Australia, 27°50'S; 118E, shallow water	Min. 19	35.5	0.3	Max. -0.30	1.05	1.35		
<b>BIVALVIA (MOLLUSCA)</b>								
<b>Pterimorphia Beurlen</b>								
<i>Mytilus californianus</i> Conrad	Max. 20.8	33.84	-0.46	Min. -1.40	-1.40	0	10	
California, 32°45'N; 117°20'W, intertidal	Min. 12.5	33.60	-0.57	Max. 0.40	0.55	0.15		
<i>Perna canaliculus</i> (Marty)	Max. 16.0	34.64	-0.1	-0.10	0.11	0.21	2	
New Zealand, 45°30'S; 171E, intertidal	Min. 6.4	33.98	-0.65	1.90	1.95	0.06		

\* Data sources indicated in the table: (1) J. L. Reid, Intermediate Waters of the Pacific Ocean, Johns Hopkins Press, 15 (1965); (2) Coastal Environ. Anal., Portobello Marine Biolog. Sta., New Zealand (1953-1968); (3) Univ. Calif. Scripps Inst. Oceanog. Data Report (1980-81) (in preparation); (4) M. K. Robinson, Atlas of the North Pacific Monthly Mean Temperatures and Mean Salinities of the Surface Layer (U.S. Naval Oceanographic Office, Washington, D.C., 1976); (5) J. Oliver (pers. comm.); (6) Pub. No. 700, U.S. Naval Oceanographic Office, Washington, D.C. (1967); (7) Oceanographic Atlas of the International Indian Ocean Expedition (pp. 1-531, K. Wyrki, ed., (1971)); (8) Univ. Calif. Scripps Inst. Oceanog. Data Reports SIO Refs. 60-27, 61-14, 62-11. (9) I. D. MacLeod (pers. comm.) and J. L. Reid, Deep-Sea Res., 16, 215 (1969); (10) Univ. Calif. Scripps Inst. Oceanog. Data Reports. SIO Refs. 78-5 and 78-16.



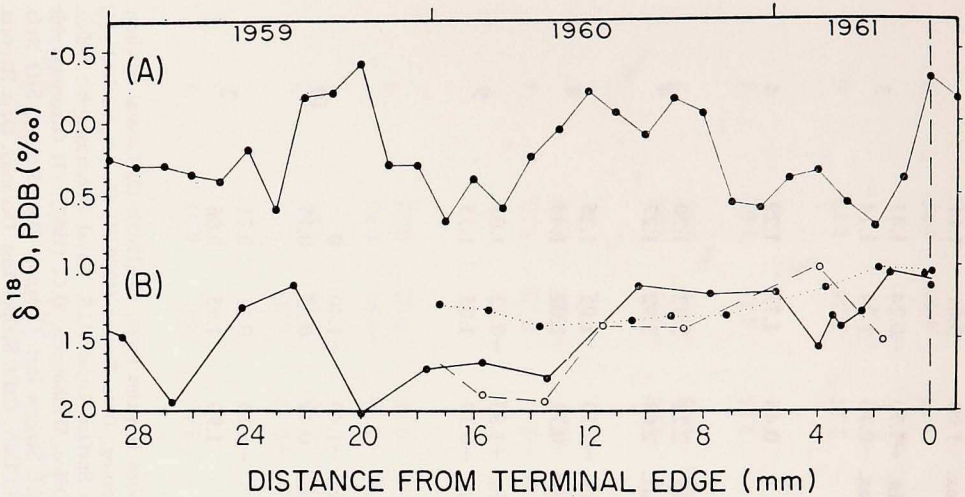


Figure 2. (A)  $\delta^{18}\text{O}$  profile calculated for equilibrium precipitation of calcite from ocean water at Monterey, California. Data points are calculated from the paleotemperature equation (Equation 1) using monthly mean temperatures and salinities recorded from 1959 to mid-1961. (B)  $\delta^{18}\text{O}$  profiles across basis (solid line); along inner sheath (dashed line) and opercular plate (dotted line) of *B. aquila* barnacle from Monterey, California (collected live in June, 1961).

Results of the X-ray diffraction analyses on the calcitic shells showed that none contained more than 1 mole %  $\text{MgCO}_3$ , therefore eliminating this possibility as a reason for the  $^{18}\text{O}$  enrichment in barnacle shells. (Mg substituted for Ca in calcite causes an increase in  $\delta^{18}\text{O}$  equal to about  $0.06\text{‰}/\text{mole } \%$   $\text{MgCO}_3$  (Tarutani *et al.*, 1969).)

The barnacle  $\delta^{18}\text{O}$  values, normalized to zero-ocean water- $\delta^{18}\text{O}$  by subtracting the corresponding ocean water  $\delta^{18}\text{O}$  estimates, are plotted against temperature in Figure 3. The mollusk curve (solid line) is that described by the paleotemperature equation of Epstein *et al.* (1953):

$$t(^{\circ}\text{C}) = 16.5 - 4.3(\delta_c - \delta_w) + 0.14(\delta_c - \delta_w)^2 \quad (1)$$

where  $t$  is the temperature,  $^{\circ}\text{C}$ , corresponding to a value of  $\delta_c$  for the  $\delta^{18}\text{O}$  value of calcite precipitated from water of isotopic composition  $\delta_w$ . (The isotopic composition reported for water is, in fact, the isotopic composition of  $\text{CO}_2$  gas equilibrated with the water at  $25^{\circ}\text{C}$  referred to  $\text{CO}_2$  liberated by acid decomposition of a standard carbonate at  $25^{\circ}\text{C}$ .) The dashed line in Figure 3 is a second order regression line fitted to the 16 barnacle data points where temperature and  $(\delta_c - \delta_w)$  values were available. (The correlation coefficient was 0.998 and the "goodness of fit" 0.997.)

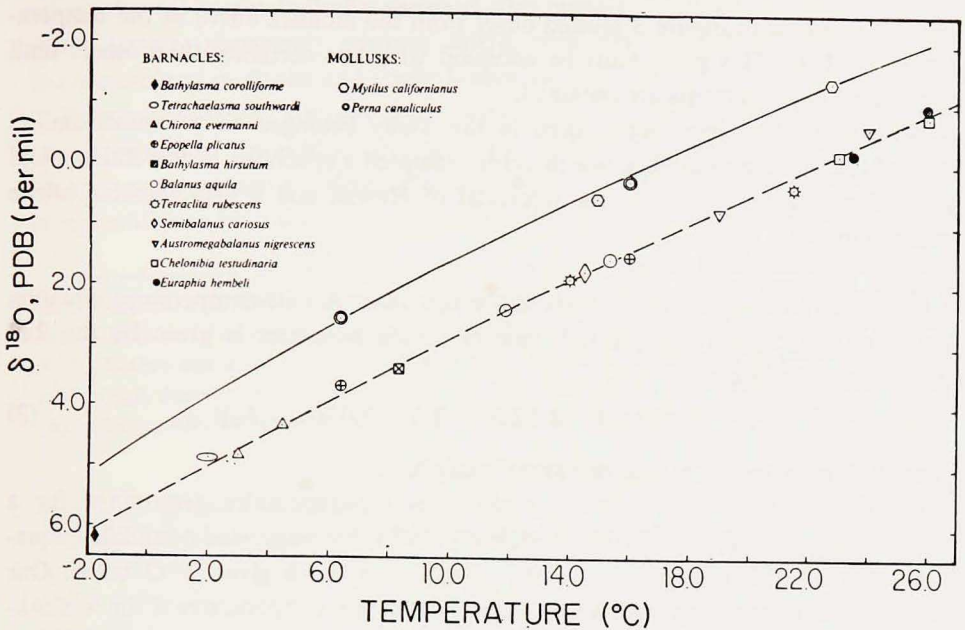


Figure 3. Comparison of the isotopic behavior of sessile barnacle calcite with temperature (dashed line) with that established for equilibrium precipitation (solid line) defined by the paleotemperature equation (Epstein, *et al.*, 1953).

The two pairs of data points on the mollusk curve (Fig. 3) were obtained from isotopic profiles on bivalved mollusk species co-occurring with two of the intertidal barnacle species (*Mytilus californianus* with *Tetraclita rubescens* at San Diego, California and *Perna canaliculus* with *Epopella plicatus* at Portobello, New Zealand). The isotopic values of the mollusks fall very close to the expected mollusk equilibrium line.

A direct mollusk-barnacle comparison also was made in the case of the *Semibalanus cariosus* which was attached to a valve of a *Mytilus californianus*. Although a seasonal range for this barnacle could not be obtained, the  $\delta^{18}\text{O}$  of the most recent growth was compared with that of the corresponding final edge of the mussel and showed a difference of 1.29‰.

*Bathylasma corolliforme* is a deep-water sessile barnacle that had been transplanted into shallow water in McMurdo Sound (Dayton *et al.*, 1982). There is a complication in interpreting the  $\delta^{18}\text{O}$  results because, although the temperature at the McMurdo location is known ( $-1.8^\circ\text{C}$ ), variations in  $\delta_w$  caused by glacier melt-water make equilibrium  $\delta^{18}\text{O}$  values difficult to estimate. The equilibrium value (3.55‰) given in Table 1 is in fact the  $\delta^{18}\text{O}$  value of a gastropod mollusk (*Limatula*) collected from the same location. The barnacle/mollusk difference here was 1.02‰



and this is shown in Figure 3 plotted offset from the mollusk curve at the temperature of  $-1.8^{\circ}\text{C}$ . This point must be accepted with less certainty than others until more direct measurements are obtained.

Most of the barnacle species used in this study belonged to the superfamilies Balanoidea and Coronuloidea. We therefore sampled a specimen of the chthamaloid genus *Euraphia hembeli* from the intertidal of Hawaii and it also secretes calcite enriched in  $^{18}\text{O}$ .

*A balanomorph barnacle paleotemperature equation.* A paleotemperature equation describing the oxygen isotopic behavior of sessile barnacles is given by the 2nd order regression fit as:

$$t(^{\circ}\text{C}) = 22.14 - 4.37(\delta_c - \delta_w) + 0.07(\delta_c - \delta_w)^2 \quad (2)$$

where  $\delta_c$  is now the  $\delta^{18}\text{O}$  value of barnacle calcite.

The original paleotemperature equation was experimentally determined for a temperature range of  $7^{\circ}$  to  $29^{\circ}\text{C}$ . Shackleton (1974) has suggested a modified equation below  $16.9^{\circ}\text{C}$  that gives a colder temperature for a given  $\delta^{18}\text{O}$  value. Our barnacle data suggest that such a trend may exist at low temperatures if the mollusk-barnacle difference is actually constant, although more measurements are necessary in the cold region to confirm this.

Preliminary analyses show that the pedunculate or stalked barnacles *Pollicipes polymerus* and *Calantica villosa* (from the intertidal of California and New Zealand, respectively), and the newly discovered deep-sea species *Neolepas zeviniae* (Killingley *et al.*, 1980) have  $^{18}\text{O}$  compositions that appear to be intermediate between mollusks and sessile barnacles (Table 1). If this difference proves to be consistent, it may have evolutionary significance. The pedunculate barnacles which first appear in the Paleozoic, do not have as complex a shell organization as do the sessile barnacles which first appear in the late Cretaceous (Newman *et al.*, 1967; Newman *et al.*, 1969). The marked difference in  $\delta^{18}\text{O}$  between the sessile balanomorph barnacles and pedunculate barnacles as well as mollusks is likely a reflection of metabolic manipulation in the production of a more complex shell.

#### 4. Summary and conclusions

We have observed a systematic and nearly constant difference between the  $^{18}\text{O}$  values of balanomorph barnacle and mollusk shells over a wide range of temperatures. The sessile barnacles secrete calcite enriched by about 1.3‰ in  $\delta^{18}\text{O}$  compared to mollusk calcite, which is regarded as being in isotopic equilibrium with ambient seawater. The difference suggests that shell-secreting mechanisms of the two taxa are quite different; mollusks are effectively in open metabolic "exchange" with ambient water during shell formation, whereas barnacles precipitate shell calcite in

which the <sup>18</sup>O is metabolically enriched with respect to ambient seawater. Pedunculate barnacles apparently produce calcite with isotopic composition intermediate between that of mollusks and sessile barnacles.

Paleotemperature estimates from fossil shells of sessile barnacles would be approximately 5°C too cold if the fractionation difference was not taken into account. Therefore we present a paleotemperature equation for sessile barnacles that enables temperatures to be calculated from δ<sup>18</sup>O measurements on sessile barnacle calcite.

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