

MEASUREMENT OF PALEOTEMPERATURES AND TEMPERATURES OF THE UPPER CRETACEOUS OF ENGLAND, DENMARK, AND THE SOUTHEASTERN UNITED STATES

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ABSTRACT

Since the abundance of the O^{18} isotope in calcium carbonate varies with the temperature at which it is deposited from water, the variation in abundance can be used as a thermometer. This paper discusses the following problems: (1) the magnitude of the effect expected, (2) the mass spectrometer of high sensitivity, (3) the preservation of the record during geological time, (4) the constancy of the isotopic composition of the ocean, (5) the impossibility of using skeletons of air breathing animals, and (6) the temperature of marine animals relative to their surroundings.

A Jurassic belemnite is used to show that the record has been retained since Jurassic times, and belemnites of the Upper Cretaceous of the United States, England, and Denmark are used to determine the temperature of this time at these localities. The temperatures are about 15° - 16° C and indicate nearly uniform temperature over this latitudinal belt. Because of the possible variation in O^{18} content of the oceans and the limited number of samples, these temperatures are regarded as preliminary.

CONTENTS

	Page
Introduction.....	399
Acknowledgments.....	400
Fractionation of isotopes in chemical processes.....	400
Problem of measuring paleotemperatures.....	401
Measurements of isotope composition, and temperatures of the Upper Cretaceous.....	406
Discussion of the North American temperatures.....	414
References cited.....	416
Addendum.....	416

ILLUSTRATIONS

Figure	Page
1. Cretaceous sections for localities studied...	413
Plate	Facing page
1. Seasonal growth of a Jurassic belemnite...	406

INTRODUCTION

Geologists have drawn many conclusions from the purely qualitative evidence of geological studies in regard to the past climatic conditions on the earth. These deductions are based upon a great variety of evidence, and the ability of the geologists to deduce as much as they have in regard to these conditions excites the wonder and admiration of all the uninitiated who examine their work even casually. Palm trees do not grow in Spitzbergen, according to all our evidence, unless the weather is mild—about the same as that of Florida. Heavy shells are deposited in warm water, and thin shells in cold water. Tree rings are definite evidence of seasons.

During the last 15 years a number of people have studied chemical properties of isotopic

substances. These studies include the demonstration of differences in the chemical properties of the hydrogen isotopes, the first calculations of such differences in the case of other of the lighter isotopes, extensive applications of such differences to the separation of isotopes and considerable evidence of such differences from surveys of the abundances of the lighter isotopes in nature. From the standpoint of the natural abundances the early demonstration that deuterium was not constant over the surface of the earth, the demonstration by Dole (1935, p. 2731; 1936, p. 268) that oxygen also varies, particularly in the air, and the variation of carbon from inorganic and organic sources by Nier (Nier and Gulbranson, 1939, p. 697; Murphy and Nier, 1941, p. 771) contributed to geology. Recently Thode, MacNamara, and Collins (1949, p. 361) have dem-

onstrated substantial variations of the sulfur isotopic abundances, and Thode, MacNamara, Lossing, and Collins (1948, p. 3008) have demonstrated such variations in boron. The present paper presents a way in which the natural variations in the abundances of the lighter isotopes makes possible, in principle at least, quantitative measurement of paleotemperatures.

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FRACTIONATION OF ISOTOPES IN CHEMICAL PROCESSES

Differences in the chemical properties of the elements, such as differences in vapor pressures and in equilibrium constants of exchange reactions, are at least partially responsible for these variations in the lighter isotopes in nature. Quite certainly the variation of the hydrogen and oxygen isotopes in the fresh waters of the earth as compared with the sea is due to the difference in vapor pressures of the varieties of water containing hydrogen and deuterium or the three oxygen isotopes, oxygen 16, 17, and 18. The variation in the abundance of the carbon isotopes discovered by Nier cannot certainly be assigned to this cause. He found that carbon 13 exists in organic matter in a decreased abundance as compared with the carbon in carbonates. Whether the photosynthetic process concentrates the carbon isotopes by some equilibrium or near-equilibrium process is not certain. Since the oxygen isotopes of the air are not in equilibrium with the oxygen isotopes of the sea or fresh water, this effect is probably also produced by the photosynthetic process or by the oxidation of organic matter by atmospheric oxygen,¹ and if a vital

¹ However, Dole has recently proposed other explanations for this effect (1949, p. 77).

selection of the isotopes occurs in the case of one of these isotopes it may well do so in others.

Variation in isotopic abundances which are due to differences in thermodynamic properties—*i. e.*, processes which are in equilibrium or

TABLE 1.—CALCULATED FRACTIONATION FACTORS FOR O¹⁶ AND O¹⁸*

<i>t</i> °C	CO ₂ - H ₂ O(<i>l</i>)	SO ₂ - H ₂ O(<i>l</i>)	PO ₄ - H ₂ O(<i>l</i>)
0	1.0220	1.0204	1.0104
25	1.0176	1.0157	1.0087
Ratio	1.0044	1.0047	1.0017
<i>K</i> ₀ / <i>K</i> ₂₅			

* McCrea (Doctoral Dissertation, Chicago, 1949) by attempting to include the effects of the potential fields in crystals instead of assuming no such fields as was done by Urey (1947) secures slightly different values for the CO₂ - H₂O constants.

very close to equilibrium—are understood in principle and in many cases in detail. On the one hand, it is necessary to know the quantum states of these molecules, and on the other it is necessary to have the exact theories of statistical mechanics in order to relate these quantum states to the physical chemical properties. For molecules in the neighborhood of ordinary temperatures and above, it is unnecessary to consider the rotational quantum states, since these are very close to classical at these temperatures in the case of all molecules except those involving the hydrogen isotopes. In this case the entire difference in the chemical properties resides in the differences in the vibrational energy levels. A great many calculations covering exchange reactions of the lighter elements were made by Urey and Greffi (1935, p. 321) and have been recalculated by Urey (1947, p. 562), together with the mathematical formulae and spectroscopic data by which the calculations are made. In general differences of a few per cent in the ratio of isotopes in two chemical compounds which are brought to equilibrium with each other with respect to the exchange of isotopes are possible for elements even as high in the periodic system as chlorine. The heavier elements can be expected to show but very slight effects of this kind. The calculations indicate that there

should be differences in the ratio of the oxygen isotopes in water and carbonate ion, water and sulfate ion, and water and phosphate ion, (Table 1).

In these calculations the results are reported for equilibrium between liquid water and the carbonate ion rather than between the gaseous water and the carbonate ion. A slight difference of a few per cent is expected in the isotopic composition, and slight temperature coefficients are predicted. The calculations show that this difference for the carbonate-water equilibrium should amount to about 0.44 per cent in 25°C, or 0.176‰ per degree C. The calculations are not exact because experimental data do not supply us with the vibrational energy levels for those molecules containing oxygen 18, and hence the vibrational frequencies of these molecules must be calculated from the known vibrational frequencies of the more abundant oxygen 16 varieties. The calculation depends upon a choice of constants. In fact, in the case of the carbonate ion five constants are needed to describe the potential energy function to the precision of quadratic terms in the co-ordinates, whereas only four independent frequencies are available for the calculation of these constants from the observed vibrational frequencies of the CO_3^{2-} ion. A similar situation exists in the phosphate and sulfate ions.

If a relation exists between the oxygen abundances in these compounds, and temperature, and the water in which the compounds were deposited, it should be possible to set up a thermometer for present-day organisms living in the sea, if the isotopic composition of the water is known not to differ from the mean of the present seas, or, in case it does, if both the isotopic composition of the carbonate and water are determined. Thus it would only be necessary to establish an empirical temperature scale and to measure the isotopic abundances of oxygen in a shell deposited by an animal in the sea in order to determine the temperature at which deposition took place. This application may be of interest in some cases, as for example in determining the temperature history of an animal, but the process which we describe is difficult, and wherever possible ordinary thermometers are likely to be considerably more convenient. On the other hand, these

effects may be preserved during geological time, and hence this method affords a possible method for measuring paleotemperatures.

PROBLEM OF MEASURING PALEOTEMPERATURES

Since the predicted temperature coefficient for the ratio of oxygen isotopes in calcium carbonate relative to water is small, mass spectrometers of very great precision and sensitivity are needed for this problem. It would be desirable to measure paleotemperatures within less than a degree Centigrade, which corresponds to a change in the atomic weight of oxygen in the calcium carbonate of 7×10^{-7} atomic weight units, or a change in the abundance of oxygen 18 by 1 part in 2.5 million of oxygen. We have set 0.01 per cent or better, corresponding to about 0.5°C or 1°F, as our objective in these experiments. As measurements of the ratio of isotopes have been conducted in the past, reports in the literature of 0.1 per cent or 1‰ are about as high a precision as has been attempted. Hence the first problem in the application of this method to paleotemperatures is the construction and operation of very sensitive mass spectrometers.

Since we have examples of animals and plants laying down isotopes out of equilibrium with their surroundings, as for example the carbon of organic material, we may well ask whether the deposition of calcium carbonate by an animal or a plant occurs so as to leave the oxygen isotopes in equilibrium with the water from which deposition takes place; that is, we may ask whether there is a *vital effect*. Probably the equilibrium deposition should be closely approximated in many cases, for the calcium carbonate deposited is constantly bathed with water from the surroundings and hence may exchange its oxygen with the surrounding water during the deposition process. However, one could quite readily imagine mechanisms which would involve the deposition of organic oxygen and thus invalidate these assumptions. The whole question could be answered only by experiment, so far as we can judge from the present state of our knowledge of the physiology of deposition of such substances. The question of deposition of phosphate on the other hand would appear to be

even more doubtful than the deposition of carbonate, since phosphate ion does not exchange its oxygen with water in which it is dissolved, and hence perhaps the phosphate deposited by animals generally does not have its oxygen in equilibrium with its surroundings. Since phosphate is so intimately involved in biochemical reactions, its oxygen may be brought to equilibrium with water by them. Only careful experimentation can determine the feasibility of the phosphate thermometer.²

Animals rarely deposit sulfate, and it is not an important substance for investigations of the type we envisage immediately. Silica is often deposited by plants and animals as a hydrous silica. Since the relative proportions of water and silica might change the relative abundance of the oxygen isotopes, this does not appear to be a very promising line of attack, though again experiments should be made to determine whether there is any possibility of using a silicate thermometer. Of the possible thermometers—that is, the carbonate, phosphate, sulfate, and silica the carbonate thermometer seems the most likely to give satisfactory results. A combination of the carbonate and phosphate would eliminate the water, and hence the isotopic variation of water always present in brackish water and fresh water would be eliminated. Dr. Martin Steinberg is investigating the possibilities of this thermometer.

Assuming that the shell is laid down in equilibrium with the water, the next important question is whether this composition is preserved during geological time. In the first place, if radical metamorphism takes place, as for example the conversion of limestone to dolomite, no satisfactory record of the isotopic abundance can be expected, for the dissolved carbonate should exchange with the water in which it is dissolved and then be redeposited with the composition partially dependent on its original composition and partially on the water which was the means of redeposition.

² Dr. Mildred Cohen (private communication) informs us that she has found that some enzymatic hydrolyses of phosphoric esters split the oxygen phosphorus bond though others do not. Hence a mechanism is available which may establish equilibrium between water and phosphate in animal bodies.

Also, if aragonite is converted to calcite through solution in water, the record would be destroyed, but, if recrystallization without the possibility of exchange occurs, the record might be preserved.

The second important method for destruction of the temperature record is diffusion through the solid state. Shells as usually deposited appear to be fairly compact, but microscopic examination shows that they consist mostly of very small crystals, often with definite canals through the shell, as for example in the case of the brachiopods or the echinoderms. Thus in considering the problem of diffusion it is not the gross size of the shell that is important, but rather the small microscopic particles, since in the course of geological time water would probably pass through the shell by hydrodynamic flow, or diffusion in the liquid would bring the isotopic composition of water within the shell to that of water in which it is immersed. The diffusion problem for considering this situation exactly would be very complicated. Assuming that the initial conditions are that (1) at the surface of the crystals the composition is that required for equilibrium with the surrounding water, and that it remains so throughout the diffusion process, and (2) that initially the composition of the crystal differed from that which would be in equilibrium with the water in which it has been immersed during the time since the shell was formed, and that it changes from this initial condition in accordance with the diffusion laws at some constant temperature, a calculation can be made which relates the size of the crystal, the time, and the coefficient of diffusion to the composition of the shell remaining. The fraction of the excess mean concentration over that in equilibrium with the surrounding water, assuming that the coefficient of diffusion is the same in all directions, is

$$\frac{Q}{Q_0} = \frac{512}{\pi^6} \left\{ \sum_{m=0}^{\infty} \frac{1}{(2m+1)^2} e^{-a(2m+1)^2} \right\}^3,$$

where $a = \frac{D\pi^2 t}{l^2}$, D is the diffusion coefficient, t the time, and l the length of the edge of the cube.

If we require that the crystal shall retain its

original composition within some arbitrary percentage, the value of $\frac{D\pi^2t}{\rho}$ can be estimated from this equation. (If one uses different values of D for different axes, the result is the same as though different lengths were used for the

TABLE 2.—VALUES OF Q/Q_0 FOR VALUES OF a^*

$a = \frac{D\pi^2t}{\rho}$	$\frac{Q}{Q_0}$
0.1	0.461
0.05	0.590
0.01	0.795
0.005	0.854
0.001	0.915
0.0001	0.964
0.00001	0.972

* We are indebted to Mr. L. G. Montet for the numerical results given in this table.

direction parallel to the axis and for the two directions perpendicular to this.)

Table 2 gives a few values of a for a few values of Q/Q_0 .

The coefficient of diffusion of one ion can be calculated from the electrical conductivity providing the current is carried by that ion and not by the other. Joffe (1923, p. 461) concluded that only the calcium ion carries the current, but to what extent the carbonate ion carries current was not determined except that its contribution was not detected. If we assume that the carbonate ion carries some fraction, δ , of the current, the coefficient of diffusion of the carbonate ion can be calculated from the resistivity of calcite from the equation,

$$D = \frac{RT\delta}{n^2F^2C\rho}$$

where n is the number of charges on the ion—*i.e.*, 2—, F is the faraday, 9650 EMU, C is the number of gram molecules of carbonate ions per cm^3 , ρ is the specific resistance in absolute units, and δ is the unknown fraction of the conductivity due to carbonate and assumed to be 0.01 here. If ρ is in ohms, this formula must be multiplied by 10^{-9} . Curie (1889, p. 385)³ measured the specific resistance of calcite,

³ See Landolt-Bornstein Tabellen II, 1062 (1923).

and his values and the values of D calculated from them are given in Table 3.

Using the first two values of $D_{\text{CO}_3^{--}}$ in the table, we can estimate the time required for diffusion to destroy the record. Thus at 20°C a crystal of 1 mm. dimensions will retain 96.4

TABLE 3.—MEASURED SPECIFIC RESISTANCE OF CALCITE AND CALCULATED COEFFICIENTS OF DIFFUSION

	$^{\circ}\text{C}$	ρ	$D_{\text{CO}_3^{--}}$
axis	20	5.5×10^{14}	4.4×10^{-23}
	100	5×10^{11}	4.8×10^{-20}
	160	3×10^{10}	8.1×10^{-19}
⊥ axis	15	9.5×10^{15}	2.5×10^{-24}
	100	2.4×10^{12}	1.0×10^{-20}
	150	1.3×10^{11}	1.9×10^{-19}

per cent of the original concentration relative to the equilibrium concentration of a changed environment for 2.3×10^{15} seconds (7×10^8 years), while at 100°C the same crystal will retain this record only 2.1×10^{11} seconds (6.4×10^4 years). Crystals of this size will not lose the temperature record due to diffusion in times sufficiently long to be of considerable interest, and the coefficients of diffusion may be even smaller than those given in Table 3.

A shell may grow a centimeter per year or 3×10^{-3} cm. per day within an order of magnitude. For simplicity we assume that the concentration of O^{18} with distance due to daily variation of temperature varies according to a cosine law. Then the diffusion equation gives

$$C = e^{-D\pi^2t/l^2} \cos \pi \frac{x}{l}$$

where we take the mean concentration as zero. If we can detect a concentration difference equal to 0.5 of the original difference between maximum and minimum, *i.e.*, at $x = 0$ and $x = l$, we have

$$\ln 2 = \frac{D\pi^2t}{\rho}$$

Substituting $D = 4.4 \times 10^{-23}$ and $l = 3 \times 10^{-3}$, the time becomes 1.4×10^{16} or half a billion years. If we ask for 0.75 of the original

variation the time becomes fifty million years. Thus even a daily variation of temperature might be partially preserved.

There is the question of whether the isotopic composition of the ocean has remained constant during geological time. Such a change would be produced, if igneous rocks of one isotopic composition are weathered and sedimentary rocks of another composition are produced. At high temperatures the isotopic compositions of all chemical substances should be the same, and, if this temperature was that of molten lava or higher, differences in isotopic composition of water and silicates should not be large and at least they should be less than at lower temperatures. The calculations referred to above show that O^{18} concentrates preferentially in the solid phases and not in the water, and it can be expected that this will be true with respect to the silicate rocks. Thus O^{18} should enter the sedimentary rocks and its concentration in the oceans should decrease with time. Assuming that the ratio of concentration in water and silicates is the same as for sulfate and water at $25^{\circ}C$, namely, 1.016, and that 600 g. of rock have been eroded for each 1000 g. of water (Goldschmidt, 1933, p. 112), it is easily shown that the water should have changed its ratio of oxygen isotopes by 3.8‰ due to this cause. Dr. P. Baertschi has shown that there are differences between ocean water on the one hand and the Disco Island Greenland basalt containing native nickel-iron and other igneous rocks, and, on the other hand, sedimentary rocks of just the kind and magnitude predicted, though the total difference between ocean water and sedimentary rocks is not 16‰ as predicted. These results will be published elsewhere.

It is of interest to inquire as to the time at which these changes took place. The carbonate equilibrium should have taken place early in the earth's history, and largely since then no important change in the O^{18} content of carbonates occurred. The shales which make up much of the sedimentary rocks should exchange their oxygen with water less readily than the carbonates, but no information is available. Baertschi finds that sandstones have an increased O^{18} content relative to the Greenland basalt and thus may have exchanged their oxygen

to some extent, though the difference is small enough to be explained by equilibrium differences in concentrations between the silica and other phases of igneous rocks. Perhaps it is a conclusion based on little more than prejudice, but we believe that it is most likely that the major part of this change occurred before the Cambrian. Until more data are available we will make no correction for this effect.

If the water of the oceans has been steadily produced during the earth's history from plutonic activity, as maintained by some (W. W. Rubey, private communication), the entire problem of the isotopic composition of the oceans must be considered from this point of view and must involve measurements on the O^{18} and D concentrations in waters which might be expected to be juvenile, as from geysers, hot springs, and volcanoes. Such measurements have not been made, but are desirable from the standpoint of the work presented here as well as for the evidence they might give relative to this suggestion in regard to the origin of the earth's water.

The oxygen of the air has a higher isotopic composition than that of the sea and fresh water. The difference in isotopic composition amounts to about 2 per cent for air oxygen as compared with the sea. All animals use atmospheric oxygen, and hence produce water that contains an increased concentration of oxygen 18. In the case of marine animals having gills which exchange water between the sea and their blood stream, it can be expected that in spite of the oxidation processes taking place within their bodies the water content of their bodies will be very close to that of the sea. One can reason that this must be true, for the water oxygen must pass through the gills approximately as readily as does the elementary oxygen, and since the amount of water present at the surface of the gills is larger than the amount of oxygen by a factor of about 10^8 , the rate of transfer of the water oxygen must be much more rapid than the transfer of elementary oxygen, and hence the isotopic composition of the blood stream must remain close to that of the sea. On the other hand, either marine or land animals that breathe air are suspect for our purposes, since atmospheric oxygen is high in oxygen 18 (*i.e.*, plus $\sim 20\%$). Land animals

drink fresh water with a decreased concentration of oxygen 18, and animals of the sea eat food containing a very considerable amount of water of the composition of water of the sea. Without knowing the relative importance of these factors it is difficult to have any confidence in any measurements made on air-breathing marine animals, and even less on land animals. It would be possible to investigate the blood of marine reptiles or mammals to determine how nearly the oxygen 18 content of their blood agrees with that of the sea. This has not yet been done.

The question arises as to whether marine animals maintain their body temperature above that of the surrounding sea. They do not. The source of heat is the combustion of food by oxygen received through the membranes of gills, usually, or of the general body surface. The membrane through which the oxygen diffuses is the same as that through which heat is lost to the surroundings. The transport of oxygen determines the rate of heat production, and this must equal the rate of loss by thermal conduction. Assuming the maximum concentration of oxygen in sea water—*i.e.*, 6 ml. per liter—and that the oxygen combines with sugar to produce 56,000 calories per gram atom of oxygen, and knowing the coefficient of diffusion of oxygen in water and the coefficient of conduction of heat in water, we can set up the following equation:

$$56,000D \frac{\Delta C}{\Delta l} = K \frac{\Delta t}{\Delta l},$$

where D is the coefficient of diffusion, K the coefficient of thermal conduction, ΔC the concentration difference of oxygen between the blood stream and the water and not greater than that corresponding to 6 ml. per liter, or .0005 gram atoms per cm^3 , Δt the difference in temperature between the water and blood stream, and Δl the membrane thickness through which both diffusion and heat conduction take place. The quantity Δl is unknown, but is the same for both processes and can be cancelled from the equation and the equation solved for Δt . Substituting $D = 2 \times 10^{-5} \text{ cm}^2\text{sec}^{-1}$ and $K = 1.42 \times 10^{-3} \text{ cal. cm. sec}^{-1}\text{ }^\circ\text{C}$, we find that $\Delta t = 0.38^\circ\text{C}$. This is the maximum difference in temperature possible under normal aerobic

conditions. Animals can expend energy for limited periods of time under anaerobic conditions and thus raise their temperature above that calculated here, but such periods would seem to be negligible.

The preparation of carbon dioxide gas for use in the mass spectrometer has constituted one of the most difficult problems in this research. It is so intimately related to the problem of the empirical temperature scale that both these problems and related ones will be discussed in a separate paper. Impurities in the carbon dioxide must not produce peaks at masses 44 and 46 such that the ratio of intensities of these masses is changed by more than one part in 10^4 , which means an impurity in mass 44 of less than this amount and in mass 46 of less than one in 2.5×10^6 . We are fortunate in using a relatively clean part of the mass spectrum. The carbon dioxide is prepared from calcium carbonate by the action of orthophosphoric acid to which phosphorus pentoxide is added to bring the concentration to approximately 100 per cent. To this enough chromic oxide is added to give a slight permanent red tint, and the excess oxide is reduced with hydrogen peroxide and the excess decomposed by heating. The calcium carbonate sample is first heated in a stream of carbon dioxide-free helium for 15 minutes at approximately 475°C in order to destroy organic matter. Approximately 9 cc. of the phosphoric acid and 50 mg. of the calcium carbonate are put in separate arms of a reaction vessel, evacuated, and brought to 25°C in a thermostat. The acid is then poured onto the carbonate. A vigorous evolution of carbon dioxide results, sometimes with a little froth which breaks during half an hour in the thermostat. The gas is then transferred to sample tubes and analyzed on the mass spectrometer. All stop cocks and tapered joints are greased with Apiezon *N* cock grease. We have never detected any effect of impurities due to this material.

With this procedure reproducible analyses are secured for both recent specimens and fossils. Using the shells of present-day marine animals grown at known temperature and correcting for the variation of isotopic composition of the various sea waters, the following equa-

tion for temperature is secured (Epstein, Buchsbaum, Lowenstam, and Urey, 1950):

$$t = 11.88 - 5.91\delta,$$

where t is measured in °C, and δ is the difference in ‰ between the ratio of O¹⁸ to O¹⁶ in a standard working gas, and this ratio for the sample, *i.e.*,

$$\delta = \left(\frac{R_{\text{sample}}}{R_{\text{standard}}} - 1 \right) 1000,$$

where R_{sample} and R_{standard} are the ratios of CO¹⁸O¹⁸ to CO₂¹⁶ for the sample and reference gas respectively. The reference gas is carbon dioxide prepared from *Belemnitella americana* from the Peedee formation of South Carolina. The slope of this curve agrees within the limits of error with that secured by McCrea (1949; 1950, p. 849) from inorganic crystals of calcium carbonate grown at constant temperature. This equation is believed to be accurate to ±1°C if the calcium carbonate were deposited in sea water of the mean isotopic composition of sea water of salinity 34.8‰. If the calcium carbonate was deposited in water of other isotopic composition, appropriate corrections must be made. The maximum variation in temperature for fully marine conditions due to the variation of isotopic composition of the oceans is ±4.5°C.

The variation of isotopic composition of the present oceans, as discussed by Epstein, Buchsbaum, Lowenstam, and Urey (1951), presents the greatest difficulty so far encountered in the measurements of paleotemperatures. These authors point out that isotopic composition varies approximately linearly with salinity. Their data on surface waters are not very extensive, but this rough linear relation applies also to surface waters. The isotopic composition varies in a rough linear way with the temperature of surface waters. This indicates that, if no correction for the isotopic composition were made, a temperature of 30°C would be read as about 25°C, while a temperature of 10°C would be read as 15°C. High temperatures would be read too low, and low temperatures too high. However, no correction to past temperature determina-

tions can be made by use of their curve because (1) A general average change in the earth's temperature would not change the average isotopic composition of the oceans, and hence a measured isotopic composition could not be corrected with their curve. (2) The slope of the curve of O¹⁸ composition vs. temperature is due to evaporation and condensation of water, and hence can be expected to be greater in times of high precipitation and lower in those of low precipitation. In general one would expect that high rates of evaporation and precipitation would accompany higher average temperatures of the earth's surface. However, we have the phenomena of glacial periods which somehow appear between limits of extremely low and extremely high temperatures, and these make estimates in regard to the relation of temperature to precipitation very difficult. For the present we do not attempt to estimate the correction for isotopic variations in the sea, but this problem is being studied from several angles and it is hoped that corrections can be applied in the future.

MEASUREMENTS OF ISOTOPIC COMPOSITION, AND TEMPERATURES OF THE UPPER CRETACEOUS

In considering geological specimens for our first attempts at measuring these temperatures, we have been guided by our considerations of probable diffusion of the carbonate in solid calcium carbonate and the improbability that recrystallized carbonates would retain the record. Since aragonite is unstable and commonly recrystallized, all recrystallized aragonitic shells will probably lose the record.

A study of fossil carbonate shells indicated that the belemnites would be the most likely to retain the record. These have a very compact structure with calcite crystals megascopically visible and so closely fitted together that internal reflections do not occur since the specimens are characteristically translucent. The polarizing microscope shows that the calcite crystals are arranged with their axes perpendicular to the axis of the guard. The orientation

PLATE 1.—SEASONAL GROWTH OF JURASSIC BELEMNITE

FIGURE 1.—CROSS SECTION OF A JURASSIC BELEMNITE

Showing relation of samples to growth rings. *W* and *S* refer to winter and summer regions respectively.

FIGURE 2.—VARIATION OF TEMPERATURE AND C¹³ CONTENT WITH RADIUS OF JURASSIC BELEMNITE

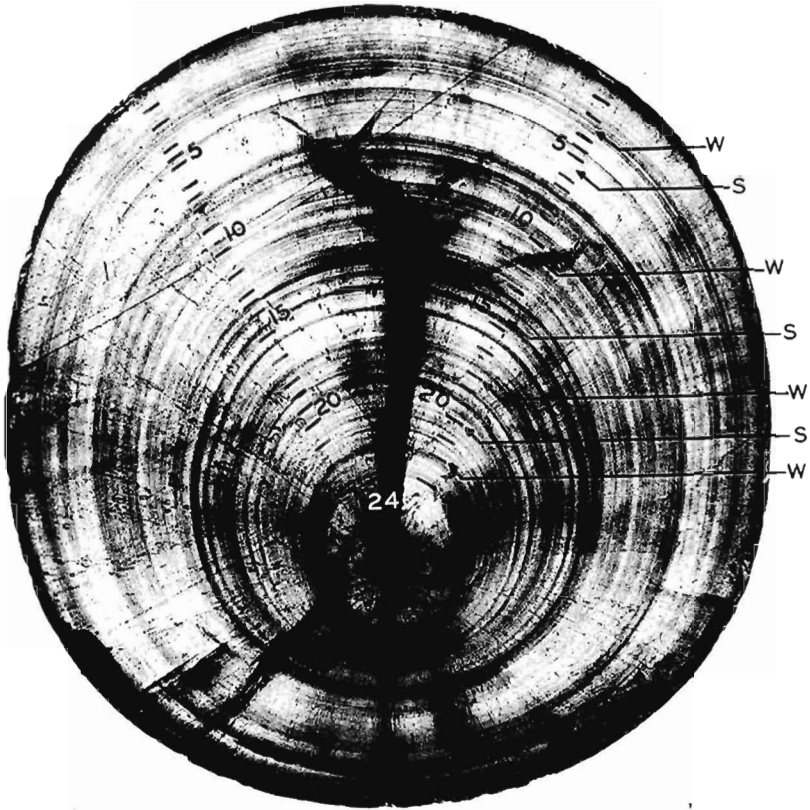


FIGURE 1

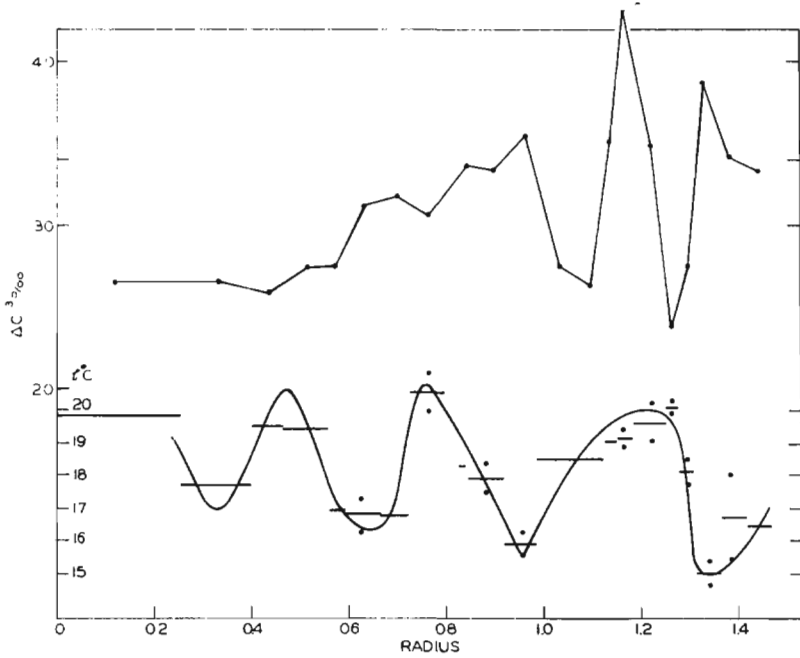


FIGURE 2

of the crystal axes and the shape and size of the crystals are evident.

Several paleontologists advised investigation of the possibility that various representative species of the genus *Inoceramus* might retain

respect to carbon and oxygen. A flat disk about 3 mm. thick was cut from the specimen, polished, and photographed in transmitted light (Pl. 1, fig. 1). Samples of carbonate were ground from the specimen following the growth

TABLE 4.—ANALYSES OF A JURASSIC BELEMNITE

Ring No.	$\delta(C^{13})$			$\delta(O^{18})$			t°C
	R	L	Av.	R	L	Av.	
1	3.3 ₄	2.5 ₀		-0.6 ₆	-1.8 ₄	-0.6 ₅ (?)	15.7
2	3.5 ₇	3.3 ₅	3.4 ₁	-0.4 ₇	-0.9 ₅	-0.7 ₁	16.1
3	3.8 ₂	3.9 ₄	3.8 ₈	-0.4 ₆	-0.3 ₀	-0.3 ₈	14.1
4	2.6 ₅	2.8 ₈	2.7 ₆	-1.0 ₄	-0.8 ₉	-0.9 ₆	17.5
5	2.4 ₁	2.3 ₇	2.3 ₉	-1.3 ₂	-1.3 ₇	-1.3 ₅	19.9
6	3.2 ₇	3.6 ₉	3.4 ₉	-1.3 ₇	-1.1 ₅	-1.2 ₆	19.3
7	4.2 ₈	4.3 ₉	4.3 ₂	-1.2 ₁	-1.1 ₄	-1.1 ₇	18.8
8	3.5 ₂	—	3.5 ₂	-1.1 ₄	—	-1.1 ₄	18.6
9	2.6 ₅	—	2.6 ₅	-1.0 ₄	—	-1.0 ₄	18.0
10	—	2.7 ₅	2.7 ₅	—	-1.0 ₅	-1.0 ₅	18.1
11	3.4 ₆	3.6 ₂	3.5 ₄	-0.6 ₂	-0.4 ₉	-0.5 ₅	15.1
12	3.2 ₉	3.4 ₀	3.3 ₄	-1.0 ₅	-0.8 ₅	-0.9 ₄	17.4
13		3.3 ₇	3.3 ₇		-0.8 ₉	-0.8 ₉	17.1
14		—	—		—	—	—
15	3.0 ₆	3.0 ₈	3.0 ₇	-1.5 ₄	-1.3 ₁	-1.4 ₂	20.3
16		3.1 ₈	3.1 ₈		-0.7 ₁	-0.7 ₁	16.1
17	3.0 ₉	3.1 ₆	3.1 ₂	-0.8 ₃	-0.6 ₄	-0.7 ₃	16.2
18		2.7 ₅	2.7 ₅		-0.7 ₅	-0.7 ₅	16.3
19, 20		2.7 ₄	2.7 ₄		-1.2 ₁	-1.2 ₁	19.0
21		2.5 ₉	2.5 ₉		-1.2 ₄	-1.2 ₄	19.2
22, 23		2.6 ₆	2.6 ₆		-0.8 ₉	-0.8 ₉	17.1
24		2.6 ₅	2.6 ₅		-1.3 ₀	-1.3 ₀	19.6
						Av. 17.6	

the temperature record. These fossils show evidence of massive calcite crystals of substantial size, though they are opaque. The calcitic layer of the shell is also present in many present-day mollusks, though usually not developed to the remarkable extent that it is in this extinct group. Experiments in progress confirm our expectation that this group retains the temperature record.

In the belemnites, a particularly convincing test for the durability of the record has been made. A Jurassic belemnite⁴ approximately 2.5 cm in diameter, brownish and translucent, with very well-defined growth rings, has been investigated for a seasonal effect and bilateral symmetry of isotopic constitution both with

lines from each side of the disk, as shown by the numbered rings shown in the photograph. In some cases right and left samples were not taken and analyzed, either because of accident in the cutting or preparation or because the samples were combined to secure sufficient carbonate for our samples. In one case both the right and left samples were lost.

The samples were analyzed for the C¹³ and O¹⁸ isotopes (Table 4).

The right and left samples of the outermost ring do not agree either in C¹³ or O¹⁸, and the check for the two samples of the second ring is rather bad for the O¹⁸. Otherwise the agreement on the two sides lies within 0.2‰. Considering probable errors of measurement, and the difficulty of cutting samples of the same thickness from the two sides, the agreement is very satisfactory. A radial variation of isotopic composition occurs both in the carbon and oxygen isotopes. The variation of the carbon isotopes does not correlate with any

⁴ Unfortunately, we do not know the species of this specimen. It was collected along with others on the Island of Skye by Dr. Cyril S. Smith, who kindly made them available to us. The specimens are of various shapes and sizes and do not appear to be a single species. As nearly as we can learn they are Jurassic, probably from the Oxford clay.

other variable found either in fossil or present-day specimens, and the variation in this specimen is of the order found in present-day specimens. The erratic variation is probably due to fractionation of the carbon isotopes by plants and the large effect that this has on the very small amount of carbon in the sea (approximately 28 mg. per liter).

Figure 2 of Plate 1 plots temperatures deduced from our O^{18} abundances against a radius measured along a line from the center of growth outward. The temperatures are plotted as horizontal lines over the range of the radius for which they are the mean temperature. The true temperature curve should intercept equal areas above and below these lines. Such a curve is drawn trying to take account of this requirement and the uncertainty of the data. Where analyses for both the right and left sides are available the line is the average, and points above and below represent the two data. The plot of δ (C^{13}) is also given and shows no correlation with the seasons. The lack of structure at the smaller radii is probably due to the fact that the samples are too thick to show the variations that may exist.

It seems difficult to believe that any "accidental" process of diffusion or exchange could have produced such a distribution of carbon and oxygen isotopes, and we conclude that the record has been substantially preserved for over a hundred million years by this belemnite. In our work on other belemnites, we have never had any reason to doubt the isotopic record providing the specimen was translucent.

This Jurassic belemnite records three summers and four winters after its youth, which was recorded by too small amounts of carbonate for investigation by our present methods; warmer water in its youth than in its old age, death in the spring, and an age of about four years. The maximum seasonal variation in temperature is about 6°C . The mean temperature was 17.6°C .

Dr. C. J. Stubblefield of the British Museum has very kindly co-operated in securing specimens of belemnites, oysters, and brachiopods from the Upper Cretaceous Chalk of England. Thus we have well-preserved specimens imbedded in a matrix which has been modified but slightly with time. Table 5 lists the speci-

mens secured from him and his record of the places and zones from which they originate.

Dr. J. Troelsen of the Mineralogical and Geological Museum of Copenhagen also supplied us with three belemnites from the lower and upper Maestrichian of Denmark. The data regarding their collection are as follows:

1. *Belemnitella mucronata*, Upper Maestrichian, 17m. below the top of the white chalk, Stevns, Denmark.
2. *Belemnitella mucronata*, Upper Maestrichian, 30-40 mm. below the top of the white chalk, Stevns, Denmark.
3. *Belemnitella mucronata*, Lower Maestrichian (white chalk), Møen, Denmark.

In studying the isotopic composition of these specimens we first removed carefully the chalk from the specimen. In the case of the belemnites the surface layers and any chalky infiltrations in cracks of the specimen were carefully removed. When possible a complete cross section of the guard was used to secure a true average temperature. In any case a piece of the solid clear calcite was ground to a fine powder and carbon dioxide prepared from these samples. In the brachiopods and oysters the chalk again was carefully removed, and parts of the clean shells were ground to fine powder and used for the preparation of the carbon dioxide samples. Whenever the chalk was available samples of the chalk in the immediate neighborhood of each specimen were analyzed. The results of the carbon and oxygen analyses for the English samples are recorded in Tables 6 and 7. In these tables the zones have been numbered as indicated in Table 5.

The oysters, brachiopods, and their chinks agree in O^{18} composition only very roughly, and their C^{13} concentrations do not agree within the limits of our measurements. The belemnites do not agree in O^{18} concentrations with their chinks nor with the oysters and brachiopods. We conclude that only the belemnites retain the temperature record. Probably the crystals of the other specimens are so small and the structure so open that diffusion has destroyed the record. In other studies now in progress we have found that a chalky belemnite—i.e., one that has lost its translucent character—agrees in isotopic composition with its surrounding chalk.

TABLE 5.—BELEMNITES, OYSTERS, AND BRACHIOPODS FROM UPPER CRETACEOUS CHALK
 All the localities are in Hampshire except the *Ostrea lunata* zone, which comes from Trimmingham. The numbers of the localities refer to *The Stratigraphy of the Chalk of Hants*, by R. M. Brydone, Publication Dulau, London 1912

Zone		Rhynchonellids	Oysters	Belemnites	
Senonian	UPPER CHALK	1. <i>Ostrea lunata</i>	Trimingham Group of " <i>Rh.</i> " <i>plicatilis</i> (J. Sowerby)	Trimingham <i>Ostrea vesicularis</i> Lamarck	Trimingham <i>Belemnitella mucronata</i>
		2. <i>Belemnitella mucronata</i>	" 1153	1153	1145
		3. <i>Actinocamax quadratus</i>	" 1086	<i>Ostrea semiplana</i> (J. de C. Sowerby) 1153	<i>Belemnitella mucronata</i> 1150
		4. <i>Offaster pilula</i>	" 1046	<i>Ostrea vesicularis</i> Lamarck 1067	<i>Actinocamax quadratus</i> 1040a
				<i>Ostrea incurva</i> Nilsson 889	<i>Actinocamax</i> sp.
		5. <i>Echinocorys scutata</i> var. <i>depressula</i>	" 918	<i>Ostrea canaliculata</i> (J. de C. Sowerby) 838	
				" 838	<i>Ostrea semiplana</i> (J. de C. Sowerby)
		6. <i>Marsupites testudinarius</i>	" 322	278	<i>Actinocamax</i> sp.
		7. <i>Uintacrinus westfalicus</i>	" 138	<i>Ostrea boucheroni</i> Coquand 138	295
		8. <i>Micraster coranguinum</i>	" 116	<i>Ostrea vesicularis</i> Lamarck " 116	<i>Actinocamax</i> sp.
9. <i>Micraster cortestudinarius</i>					
10. <i>Holaster planus</i>					
Turonian	MID-DILE CHALK	11. <i>Terebratulina lata</i>	" 116		
		12. <i>Inoceramus labiatus</i>	80 " <i>Rh.</i> " <i>cuvieri</i> d'Orbigny		
Cenomanian	LOWER CHALK	<i>Holaster subglobosus</i>	" 34	54	
		13. <i>Actinocamax plenus</i>		<i>Actinocamax plenus</i>	
		14. <i>Schloenbachia varians</i>	21 " <i>Rh.</i> " <i>mantelliana</i> (J. de C. Sowerby)		

TABLE 6.—THE QUANTITY $\delta(C^{13})$ FOR UPPER CRETACEOUS OF ENGLAND

Zone	Brachiopod		Oyster		Belemnite	
	Specimen	Chalk	Specimen	Chalk	Specimen	Chalk
1	1.3 ₉	0.9 ₄	0.7 ₉	0.7 ₁	0.4 ₆	1.6 ₄
2	2.8 ₃	1.8 ₇	2.0 ₀	1.5 ₅	2.1 ₁	1.3 ₉
3	2.5 ₃	1.6 ₁	1.8 ₀	1.7 ₆	1.1 ₂	1.3 ₂
4	2.9 ₀	—	2.2 ₆	1.7 ₂	0.9 ₂	—
5	2.4 ₀	1.8 ₀	1.6 ₈	1.1 ₉	—	—
6	2.3 ₂	1.6 ₇	1.8 ₇	1.6 ₇	1.8 ₅	—
7	—	—	—	—	—	—
8	—	1.5 ₈	1.6 ₇	1.5 ₂	—	—
9	1.5 ₃	0.7 ₅	2.0 ₉	1.0 ₄	—	—
10	1.9 ₇	1.1 ₇	1.3 ₀	0.9 ₀	—	—
11	—	—	1.2 ₂	1.2 ₂	—	—
12	—	—	—	—	—	—
13	—	—	2.8 ₄	2.8 ₁	3.2 ₀	—

Some specimens of Table 5 were not used because of bad preservation.

constant composition had been reached, all the analyses should be identical, and thus we conclude that this is not the case.⁵

The oxygen analyses for the Danish specimens together with derived temperatures are given in Table 8. The specimens are numbered as given previously.

Upper Cretaceous paleotemperature data for North America were secured from the *Exogyra costata* zone of a number of widely scattered outcrops of the Gulf Coast and Eastern Seaboard of the United States between western Tennessee and northeastern Mississippi north-eastward to New Jersey.⁶ The specimens from Tennessee were derived from the sandy marls of the basal third of the exposure on Dave Weeks' place, 3½ miles south of Enville, McNair County, the type locality of the Coon Creek tongue of the Ripley formation (Wade,

TABLE 7.—THE QUANTITY $\delta(O^{18})$ FOR UPPER CRETACEOUS OF ENGLAND

Zone	Brachiopod		Oyster		Belemnites		Belemnite Temperature °C
	Specimen	Chalk	Specimen	Chalk	Specimen	Chalk	
1	-1.4 ₆	-2.3 ₉	-1.8 ₅	-2.2 ₆	-0.5 ₅	-1.8 ₃	15.0
2	-1.9 ₃	-2.9 ₀	-2.0 ₉	-2.9 ₂	-0.4 ₂	-2.5 ₂	14.4
3	-2.4 ₀	-2.4 ₆	-2.9 ₉	-2.6 ₆	-1.2 ₄	-3.8 ₅	19.2
4	-1.9 ₅	—	-1.9 ₁	-2.6 ₆	-1.2 ₆	—	19.3
5	-2.4 ₃	-2.2 ₀	-2.7 ₃	-2.7 ₄	—	—	—
6	-2.1 ₀	-2.4 ₆	-2.4 ₄	-2.5 ₆	-2.0 ₃	—	23.8
7	—	—	—	—	—	—	—
8	—	-2.6 ₈	-2.8 ₉	-2.3 ₃	—	—	—
9	-2.8 ₃	-2.9 ₄	-2.0 ₂	-2.5 ₉	—	—	—
10	-2.6 ₀	-2.4 ₇	-3.6 ₈	-2.7 ₅	—	—	—
11	—	—	-3.6 ₂	-3.1 ₆	—	—	—
12	—	—	—	—	—	—	—
13	—	—	-1.8 ₉	-3.5 ₄	-0.4 ₀	—	14.2
14	—	—	—	—	—	—	—

Present-day specimens show marked variation in C^{13} concentrations even when they grew in the same region at the same time. Thus the samples considered here should have had identical or nearly identical O^{18} concentrations, but not identical C^{13} concentrations when first buried. If both the oxygen and carbon concentrations changed with time we would expect that the oxygen concentrations would tend to change by comparable amounts, and hence remain somewhat the same as observed. On the other hand the carbon concentrations should show no such approximate agreement. If complete equilibrium with water and carbon of

1926). Though belemnites do not occur in these deposits, depriving us of our reliable standard

⁵ We have analyzed a few samples of ooze from the Pacific and Atlantic Oceans in co-operation with W. F. Libby and J. R. Arnold, who are working on C^{14} concentrations in such materials. We are unable to interpret the results in any consistent way as yet, and evidently more data are required.

⁶ The specimens from Western Tennessee, Mississippi, North Carolina, and South Carolina were collected by H. C. Urey and H. A. Lowenstam. We are indebted to Dr. L. W. Stephenson for advising us on certain collecting localities in South Carolina, to Dr. Horace C. Richards who guided us to the New Jersey outcrops, and to Mr. P. A. Bethany of Macon, Mississippi, who advised us on and collected with us from the outcrops of the Macon area.

for comparison, the excellent shell preservation of all the molluscan elements with originally aragonitic shells, elsewhere in the outcrop areas preserved only as casts, implied exceptionally favorable burial conditions suggestive of preservation of original isotopic abundances. The specimens analyzed from northeastern Mississippi were secured from the Prairie Bluff Chalk, at two localities, the glades on the Vanny Jackson farm west of the Rocky Hill Church road, 3.6 miles north-northeast of Starkville, Octibbeha County, which is the general area of fossil locality 6846 of Stephenson and Monroe (1940), and from glades on the A. Lindley farm 4 miles southwest of Macon, Noxubee County (Stephenson and Monroe's 1940 collecting locality 17242, 17484). At both localities, the specimens were found loose in the residual fossil concentrations which littered the glades, since Prairie Bluff specimens satisfactorily preserved and located *in situ* were unavailable. The positions marked 10 feet and 13 feet above the base of the Prairie Bluff formation in Table 9 for the specimens from the Lindley locality refer to collecting levels on the glade and not to stratigraphic positions. Despite the rich assemblages in the glade concentrations, only originally pure calcitic skeletal forms on the calcitic layer of calcite and aragonitic inter-layered shells are preserved as such, reducing the number of potential elements for paleotemperature determinations to crinoidal remains, echinoids, *Terebratulina floridana*, Ostreidae, and Pectinidae among the pelecypods and guards of *Belemnitella americana*, the balance consisting of molds, prevalently in phosphatic form. Shark teeth occur with the invertebrate assemblages composed of carbonate skeletons and furnish source material for a possible future check of the carbonate thermometer by the phosphate thermometer.

The South Carolina specimens were secured *in situ* from the basal section of the Peedee formation which is well exposed in the west bank of the Peedee River at Burches Ferry, 15½ miles from Florence. This is the locality from which Stephenson (1923) described and figured a number of well-preserved specimens of *Belemnitella americana* and described a new species. At the time of our visit, the river was low enough to expose a total of 13½ feet of Pee-

dee strata, consisting of calcareous, argillaceous, glauconitic sand above 3½ feet of the shales of the Black Creek formation. *Belemnitella americana* was found in small numbers through the entire local Peedee section; it was concentrated in conspicuous numbers in a narrow 9-inch zone, 57 inches above the base of the formation.

TABLE 8.—OXYGEN ISOTOPIC ANALYSES AND TEMPERATURES OF THE DANISH BELEMNITES

Specimen	$\delta(0^{18})$	$t^{\circ}\text{C}$
1	-0.1 ₇	12.8
2	-0.4 ₃	14.3
3	-0.0 ₉	12.3

The *Belemnitella* guards are throughout oriented more or less horizontally, with occasional deviations ranging up to about 10° from the horizontal plane. Within the horizontal plane the belemnite rostra display random orientation as far as could be ascertained in the vertical exposure. *Exogyra costata* occurs sparingly distributed through the basal 7½ feet of the section, the individuals being generally small and thin shelled. Larger, but comparatively fragile individuals are abundant in the overlying 22 inches of the section which is conspicuously fossiliferous, consisting of a semicoquina of pelecypod shell debris. The specimens analyzed were taken *in situ*.

The North Carolina specimens were collected from a 6-inch calcareous sandstone ledge of the Peedee formation, exposed above water level, and 6 inches of soft marl below water level at the Jackson's Hole Landing, locality of Stephenson (1923, p. 25), Northeast Cape Fear River. A number of large, heavy-shelled *Exogyra costata* individuals, with strongly resorbed surfaces, and a single rostrum of *Belemnitella americana*, were secured *in situ* from the water-covered marly layer, which contrasts strongly with the rich mollusk assemblages of overlying calcareous sandstone ledge. Pectinidae and *Belemnitella americana*, originally calcitic skeletal types alone are well preserved; the originally aragonitic shells of the associated pelecypods is often represented by casts and molds. As the Peedee section lacks top and bottom it is not possible to determine the stratigraphic position of this section within the formational range.

TABLE 9.—OXYGEN ISOTOPIC ANALYSES AND TEMPERATURES FROM *Belemnitella americana* FROM THE SOUTHEASTERN UNITED STATES

Formation	Locality	Level	Analysis	Temp.	Av. Temp.
Prairie Bluff	Starkville, Miss. 3.6 miles N-NE. on Rocky Hill Church Road	Unknown	(1) -0.5 ₃	14.9	16.4
			(2) -0.9 ₇	17.6	
			(3) -1.2 ₃	19.1	
			(4) -1.0 ₉	18.2	
			(5) -0.1 ₁	12.5	
	Macon, Miss. A. Lindley place 4 miles SW. of Macon	13 feet above base of Prairie Bluff	(1) 0.0 ₀	11.9	11.9
Peedee	Peedee River, S. C.	105 inches above base of formation	(1) -0.5 ₁	14.9	14.9
		57 inches to 68 inches above base of formation	(1) -0.4 ₄	14.5	14.4
			(2) -0.3 ₀	13.6	
			(3) -0.2 ₈	13.4	
			(4) -0.3 ₁	13.7	
			(5) -0.2 ₇	13.4	
			(6) -0.6 ₇	15.8	
			(7) -0.5 ₅	15.1	
			(8) -0.4 ₈	14.6	
			(9) -0.2 ₈	13.5	
			(10) -0.6 ₆	15.8	
(11) -0.6 ₁	15.5				
Cape Fear River	Unknown	(1) -0.5 ₄	15.1	16.1	
		(2) -0.8 ₉	17.1		
Navesink	Mullica Hill, N. J.	Base of formation	(1) -1.2 ₁	19.1	18.0
			(2) -1.1 ₃	18.8	
			(3) -0.9 ₀	17.2	
			(4) -0.8 ₁	16.7	
			(5) -1.0 ₀	18.8	
			(6) -0.9 ₈	17.6	
	Hornerstown, N. J.	Base of formation	(1) -0.8 ₇	17.0	16.0
			(2) -0.5 ₃	15.0	
			(3) -0.4 ₉	14.8	
			(4) -0.7 ₅	16.2	
			(5) -0.8 ₀	16.6	
			(6) -0.8 ₀	16.6	
			(7) -0.3 ₉	14.2	
			(8) -0.6 ₂	15.5	
Cream Ridge	Unknown	(1) -0.5 ₀	15.4	15.7	
		(2) -0.7 ₀	16.0		

The New Jersey specimens analyzed were secured from the Navesink marl at the well-known fossil locality in the hillside just south of the railroad trestle at Mullica Hill (Weller, 1907, locality 169), in the cut of a tributary of

convexa, and *Belemnitella americana*. The dense packing of the pelecypod shells with the randomly oriented, interwedged *Belemnitellas* indicates post mortem concentrations swept together in shallow agitated waters. Burial

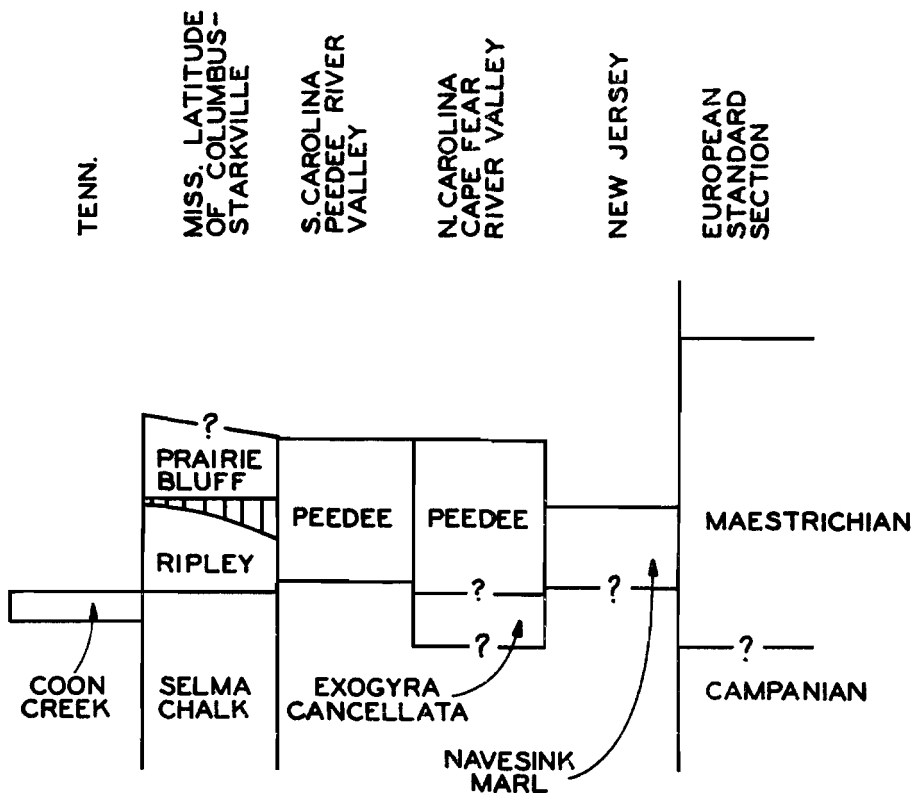


FIGURE 1.—CRETACEOUS SECTIONS FOR LOCALITIES STUDIED

Crosswicks Creek near Hornerstown, and from the dump piles of the abandoned "marl pit" on Edgar Schank's farm near Cream Ridge (Weller, 1907, locality 127). The Mullica Hill specimens were derived from a 4-inch shell bed, consisting of *Exogyra costata*, Pectinidae, and *Belemnitella americana* in a marly glauconitic sandy matrix about 12 feet above the base of the bluff exposure. The preservation of the pelecypods and belemnites is generally poor due to leaching and partial limonitic replacement, while conspecific forms occurring in shell beds, located 10 feet lower and about 3 feet higher up in the bluff section are represented only in the form of casts and molds. The Hornerstown specimens are derived from a shell bed, composed principally of *Exogyra costata*, *Gryphaea*

grounds foreign to the habitat are thus strongly implied for the *Belemnitella* element. According to Spengler and Peterson (1950), the shell beds mark the base of the Navesink member of the Monmouth formation, previously interpreted as a formation and group respectively. A basal position for the Mullica Hill and Hornerstown specimens in the Navesink is thus indicated, while the stratigraphic position of the Cream Ridge forms, secured loose from the dump piles, cannot be determined.

All the North American specimens included in the present study fall within the range of the *Exogyra costata* zone, as shown by the correlation of the formations from which they were derived (Fig. 1). In comparing the stratigraphic positions of the specimens secured from Tennes-

see, Mississippi, and South Carolina, and the Mullica Hill and Hornerstown localities in New Jersey within their respective formations and in relation to the formational ranges and their correlation, it will be seen that the Coon Creek forms, from southwestern Tennessee, occupy a lower; those from the Peedee formation of South Carolina and from the Navesink localities of New Jersey an intermediate, and the Prairie Bluff forms from northeastern Mississippi a higher position within the zonal range. Their stratigraphic relations to the North Carolina and Cream Ridge locality of New Jersey cannot be determined because of the unknown stratigraphic positions of the latter within their respective formations. To judge from the narrow range of the Navesink marl, the New Jersey specimens may be anywhere between the top of the Coon Creek formation of southwestern Tennessee and the base of the Prairie Bluff chalk in northeastern Mississippi, those from the Peedee formation of North Carolina a considerably wider interval.

The North American forms serve for an initial survey of the special paleotemperature range within the time interval occupied by the *Exogyra costata* zone, even though in some cases our specimens came from different levels within that zone and in other cases the relationships within the zone are uncertain.

DISCUSSION OF THE NORTH AMERICAN TEMPERATURES

The *Exogyra costata* zone is correlated with the Maestrichian of the European standard section (Stephenson and Reeside, 1938, p. 1637; Stephenson et al., 1942, Chart No. 9). *Belemnitella mucronata* which occupies in the Upper Senonian of northern and central Europe a stratigraphic position similar to *Belemnitella americana* in southeastern North America, has been considered closely related to the North American species (Stephenson, 1923, p. 56). Approximate time equivalency of their enclosing strata is further indicated by other faunal and floral similarities. Keeping in mind the as yet existing uncertainties in intercontinental correlation, particularly with reference to details in time equivalency and length of time intervals, it is nevertheless permissible to compare in general terms the Maestrichian paleo-

temperature data from both sides of the Atlantic that is, of the Gulf Coastal area in North America and of southern England and Denmark in northwestern Europe. There is surprisingly little difference in the temperatures recorded by the reliable forms from either side of the Atlantic despite the latitudinal range of the collecting localities, which extend from 33° to about 41°N. in North America to between about 52° and 56°N. in England and Denmark.

In surveying the meaning of the temperature records only those of the belemnites can be at present considered reliable. As the temperatures recorded constitute at best mean temperatures of skeletal growth periods, they do not necessarily represent annual mean temperatures of the waters in which they lived. This is clearly indicated by the temperature range recorded by the shell of a recent *Chama macerophylla* from the Bermuda inshore waters, which has been analyzed for seasonal effects. Shell material is laid down only after the surface waters reach 24°C and extends through the warm part of the year, which reaches a maximum at 29°C, while the yearly temperature range of the inshore waters covers a range from 18°C to 29°C. The mean temperature of 26½°C recorded by the *Chama macerophylla* shell is thus 2½°C higher than that of the waters. Organisms with a narrow temperature range for skeletal growth, particularly with a growth period largely off the average with reference to the yearly range of their waters, would constitute unreliable data for climatic records unless forms from identical habitat with wider or overlapping skeletal growth periods are available for comparison. The *Inoceramus* lineage now under investigation may furnish a comparative element for correction at a future date.

Returning to the belemnites, preference for cooler waters is strongly implied by their geographic distribution. Dacqué (1915, p. 423-426), in discussing the paleoclimatology of the Cretaceous, pointed out that broad climatic zoning is introduced in the beginning of the Cretaceous and becomes sharply established by *Upper Cretaceous* time. Differentiation into a boreal and Mediterranean-aquaetorial zones is fairly well documented, the aquaetorial zone being characterized by prominent carbonate sediments, reef-forming corals and rudistids,

large Foraminifera, and heavy-shelled Nerineidae and actaeonellids. Most convincing is the appearance of dwarfed and crippled rudistids in the bordering boreal zones both to the north and south, as shown by Dacqué's distribution map for the rudistids. *Belemnitella* was shown to be confined to the northern boreal zone of the Upper Cretaceous seas. In comparing the size ranges of the collections of *Belemnitella americana* secured from the various localities of the Gulf Coastal area, an increase in maximum sizes attained is clearly noticeable from Mississippi northeastward to New Jersey. The Mississippi localities were apparently located in the southern border area, as Stephenson and Monroe record representatives of the rudistid genus *Durania* from the *Exogyra cancellata* subzone of the upper Selma Chalk in which *Belemnitella americana* makes its first appearance.

The northeastward trend of increasing sizes in *Belemnitella americana* could be correlated with cooler water preference, but their mean temperature records do not seem to support this view. Refinement in the sampling procedure contemplated for studies of seasonal variations may decide this question. Throughout our discussion we have neglected the possibility and even probability of some variation of the isotopic composition of the waters, and this may explain the nearly identical temperatures of the Carolina and Mississippi region as contrasted to New Jersey.

The depth range is another factor yet to be considered in the evaluation of the temperatures recorded by nectonic elements such as belemnites. The sediment character and the burial assemblages of the American representatives in the locations examined indicate that, though the bottoms were largely below effective wave base (except for New Jersey examples, which occur in wave concentrated shell beds), the abundant and varied mollusk forms point to comparatively shallow bottoms, well within the euphotic zone. This agrees with Naef's (1922) conclusions of prevalent shallow-water habitat for the belemnites in general as implied by functional morphology. Thus the temperature variation should be characteristic of a limited depth range. That *Belemnitella* lived in shallow cooler waters is further indicated by the absence of stray individuals in the southern Tethys waters.

The narrow gradient indicated by the mean temperatures of the *Belemnitellas* across the latitudinal width from Mississippi to Denmark is surprising. Unless the temperature tolerance of skeletal growth of the European and American species happens to coincide, rather uniform climatic conditions are suggested over most of the boreal zone during the Maestrichian. Gradual lowering of the temperatures through Upper Cretaceous time toward the Maestrichian are indicated in England and agree with the establishing of the climatic zones noted by Dacqué. Increased mixing of the oceans due to stronger current development should have resulted. North America and South America were probably separated by the circumequatorial Tethys, and the waters from the pole area previously connected with it via the western interior strait became separated at about the critical time that the lowering of the late Cretaceous temperatures is indicated to have taken place. Free mixing of the northern waters due to lack of the present existing north-south land barriers should have tended to equalize the temperatures toward the pole. Subtropical climatic conditions could have thus been maintained on the northern land masses where the dinosaurs continued to flourish.

Cowles (1939; 1940; 1945) has suggested extinction of the dinosaurs at the closing interval of the Cretaceous due to increased temperatures. On the other hand continued lowering of the temperatures as suggested by the English downward trend would have equally caused their extinction. We do not wish to emphasize this apparent downward trend since our temperatures depend on single samples in each zone and our experience indicates that a variation between a number of specimens of the same locality of as much as 5°C is possible. The investigation of more specimens from these zones and seasonal variations of such specimens may enable us to reach a definite conclusion. Longitudinal paleotemperature traverses for the entire Upper Cretaceous extending into early Tertiary, based on a large number of associated faunal elements and including benthonic forms should furnish a clearer trend of paleoclimatologic conditions as well as aid in quantitatively evaluating the temperature niches of the marine organisms.

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ADDENDUM

It is necessary to consider the possible effect of the changed temperature scale mentioned in the Addendum of the preceding paper on the temperatures recorded here. Repetition of the analyses using the modified method of preparation and the corrected temperature scale on 10 specimens reported in this paper changed their measured temperatures by an average of 1.1°C with a spread of -0.6° to +2.2°C. The temperatures remain remarkably uniform though systematically higher by about 1°C.