

# Precise Timing of the Last Interglacial Period from Mass Spectrometric Determination of Thorium-230 in Corals

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The development of mass spectrometric techniques for determination of  $^{230}\text{Th}$  abundance has made it possible to reduce analytical errors in  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$  dating of corals even with very small samples. Samples of  $6 \times 10^8$  atoms of  $^{230}\text{Th}$  can be measured to an accuracy of  $\pm 3$  percent ( $2\sigma$ ) and  $3 \times 10^{10}$  atoms of  $^{230}\text{Th}$  can be measured to an accuracy of  $\pm 0.2$  percent. The time range over which useful age data on corals can be obtained now ranges from about 50 to about 500,000 years. For young corals, this approach may be preferable to  $^{14}\text{C}$  dating. The precision with which the age of a coral can now be determined should make it possible to critically test the Milankovitch hypothesis concerning Pleistocene climate fluctuations. Analyses of a number of corals that grew during the last interglacial period yield ages of 122,000 to 130,000 years. The ages coincide with, or slightly post-date, the summer solar insolation high at  $65^\circ\text{N}$  latitude which occurred 128,000 years ago. This supports the idea that changes in Pleistocene climate can be the result of variations in the distribution of solar insolation caused by changes in the geometry of the earth's orbit and rotation axis.

FLUCTUATIONS IN CLIMATE RESULT IN CHANGES IN THE mass of water stored as ice in continental glaciers, which in turn cause changes in sea level. Because some species of coral grow very close to the sea surface, they can be used as indicators of the height of sea level in the past. The  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$ - $^{232}\text{Th}$  system has been used to delineate the absolute chronology of events in the last 150 ky (1 ky =  $10^3$  years) and has been applied to fossil corals to establish an absolute chronology for sea level changes (1-3). This approach has been particularly important in testing the astronomical theory of climate change which was formulated by Milankovitch (4). This theory states that the fluctuations in Pleistocene climate are caused by changes in the distribution of solar energy received by the earth due to changes in (i) the obliquity of the ecliptic, (ii) the eccentricity of the earth's orbit, and (iii) precession of the earth's rotation axis. On the basis of calculations from the orbital parameters, the summer solar insolation received at  $65^\circ\text{N}$  latitude was shown to be a function of time (4) and was viewed as a forcing function controlling the earth's climate. High values of summer insolation would favor deglaciation, low values would favor glaciation. The response of sea level height to orbital forcing can be modeled from the dynamics of the ocean, glacial growth and

melting, and isostatic rebound. This is a particularly active area of research (5, 6).

The test of sea level height with time would depend on the accuracy of the orbital parameters and the accuracy of the sea level curve derived from the geologic record. If the insolation curve and the sea level curve were found to be identical, this would demonstrate that (i) the earth's climate responds to orbital forcing; (ii) the response shows no measurable phase lag; and (iii) of the possible curves that could be calculated from the orbital parameters, the summer solar insolation received at  $65^\circ\text{N}$  is the one that controls climate. Differences between the insolation curve and the sea level curve would indicate that at least one of these statements is not true.

Two approaches have been used to determine the height of sea level in the past. The ratio of  $^{18}\text{O}$  to  $^{16}\text{O}$  in seawater is a function of the fraction of water stored as ice in glaciers. Detailed measurements of  $^{18}\text{O}/^{16}\text{O}$  as a function of depth in deep sea cores have provided a continuous record of climatic change over the past  $10^6$  years (7, 8). Spectral analysis of this record has provided convincing evidence that at least some of the variability in Pleistocene climate is the result of orbital forcing. However, the approach has been limited by the inability to independently assign an absolute chronology to this record.

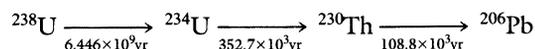
A second approach is the dating of coral terraces. This does not provide a continuous record of sea level change but has the advantage that corals of appropriate age can be dated by  $^{230}\text{Th}$  methods. An apparent correlation has been drawn between the ages of a number of coral terraces thought to have grown during periods of high sea level (interglacial and interstadial periods) and the times of high insolation (1-3). However, this approach has been limited by the analytical uncertainty of data obtainable with present methods, some discrepancies in results, and possible open-system behavior of corals with respect to uranium and thorium.

Analysis of  $^{230}\text{Th}$  and  $^{234}\text{U}$  has typically been done by  $\alpha$  spectrometry. The precision with which a measurement can be made with this technique has practical limitations resulting from sample size and counting time. On the basis of our experience in analyzing  $10^9$  to  $10^{10}$  uranium atoms by means of thermal ionization mass spectrometry (9), we have attempted to supplement the  $\alpha$ -counting method with mass spectrometric measurements as a means of increasing precision and decreasing sample size. In order to compare this

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technique with previous methods, we have analyzed a number of corals that had been dated by  $^{14}\text{C}$ ,  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$  ( $\alpha$  counting), or counting of coral growth bands. Included among these samples is a series of corals that are thought to have grown during periods of high sea level 80 to 140 ky ago. These corals were chosen for analysis since the precise ages of these samples and their relation to the insolation curve would provide a critical test of the Milankovitch theory. The  $^{238}\text{U}$  and  $^{232}\text{Th}$  concentrations in a modern mollusk shell have also been measured as a means of assessing the suitability of this material for dating.

**Systematics.**  $^{230}\text{Th}$  dating of corals is based on the decay of  $^{238}\text{U}$  through a series of relatively short-lived intermediate daughters to  $^{206}\text{Pb}$ . The pertinent nuclides in this decay chain are:



where the numbers below the arrows are mean lives. For simplicity, the other short-lived intermediate daughters in the decay scheme have not been shown. The initial amount of  $^{230}\text{Th}$  in corals can be calculated by multiplying the  $^{230}\text{Th}/^{232}\text{Th}$  ratio of surface ocean water [see (10, 11)] by the  $^{232}\text{Th}$  abundance in corals determined in our study. The calculated value is extremely low. It is equivalent to the amount of  $^{230}\text{Th}$  generated by radioactive decay in about 1 year and can be taken to be zero. If we use this initial condition and assume that corals represent closed systems, the equations for radioactive production and decay can be solved.

$$1 - [^{230}\text{Th}/^{238}\text{U}]_{\text{act}} = e^{-\lambda_{230}T} -$$

$$\left( \frac{\delta^{234}\text{U}(0)}{1000} \right) \left( \frac{\lambda_{230}}{\lambda_{230} - \lambda_{234}} \right) \left( 1 - e^{(\lambda_{234} - \lambda_{230})T} \right) \quad (1)$$

$$\delta^{234}\text{U}(T) = \delta^{234}\text{U}(0)e^{\lambda_{234}T} \quad (2)$$

Equation 1 [modified from Kaufman and Broecker (12)] gives the age ( $T$ ) as a function of the decay constants ( $\lambda$ ) and the measured  $^{234}\text{U}/^{238}\text{U}$  and  $^{230}\text{Th}/^{238}\text{U}$  ratios. Equation 2 gives the initial  $^{234}\text{U}/^{238}\text{U}$  ratio when the coral grew as a function of the measured  $^{234}\text{U}/^{238}\text{U}$  ratio and  $T$ . For convenience, the  $^{234}\text{U}/^{238}\text{U}$  ratio has been reformulated into  $\delta$  notation. The present  $\delta^{234}\text{U}$  value is

$$\delta^{234}\text{U}(0) = \{ [(^{234}\text{U}/^{238}\text{U}) / (^{234}\text{U}/^{238}\text{U})_{\text{eq}}] - 1 \} \times 10^3$$

This represents the fractional deviation of the measured  $^{234}\text{U}/^{238}\text{U}$  ratio from the  $^{234}\text{U}/^{238}\text{U}$  value at secular equilibrium [ $(^{234}\text{U}/^{238}\text{U})_{\text{eq}}$ ] in parts per thousand. The initial  $\delta^{234}\text{U}$  value when the system was isolated from seawater is given by  $\delta^{234}\text{U}(T)$ . The measured  $^{230}\text{Th}/^{238}\text{U}$  atomic ratio has been represented as an activity ratio [ $(^{230}\text{Th}/^{238}\text{U})_{\text{act}}$ ] by multiplying the  $^{230}\text{Th}/^{238}\text{U}$  atomic ratio by the ratio of the decay constants ( $\lambda_{230}/\lambda_{238}$ ).

If  $\delta^{234}\text{U}(T)$  is known, the equations represent two independent chronometers. We have chosen to use Eq. 2 not as a chronometer but as a means of calculating  $\delta^{234}\text{U}(T)$  with the value of  $T$  determined from Eq. 1. If we assume that the uranium isotopic composition of seawater is constant with time, a difference between the calculated  $\delta^{234}\text{U}(T)$  and the present seawater value (11) would indicate that the uranium in the coral was not derived via closed-system evolution from normal seawater. The calculated  $\delta^{234}\text{U}(T)$  therefore provides an independent check of whether the coral has behaved as a closed system.

$^{232}\text{Th}$  is not a nuclide in the decay chain shown above and does not appear in Eqs. 1 or 2. It has an extremely long mean life [ $2.0212 \times 10^{10}$  years (13)] and can be considered stable over the time range considered here. Comparison of the  $^{232}\text{Th}/^{238}\text{U}$  ratio in

an old coral with the  $^{232}\text{Th}/^{238}\text{U}$  ratio in modern corals would indicate whether substantial differential addition or leaching of  $^{238}\text{U}$  or  $^{232}\text{Th}$  had occurred subsequent to coral growth. Since the chemical properties of  $^{230}\text{Th}$  and  $^{232}\text{Th}$  are the same, knowledge of the  $^{232}\text{Th}/^{238}\text{U}$  ratio provides another check of the closed-system assumption used in deriving Eq. 1.

**Analytical procedures and samples.** Detailed analytical procedures for measurement of the Th and U isotopes have been described (11, 14). In Table 1, we have listed, for each analysis, the amount of coral analyzed, the number of  $^{230}\text{Th}$  atoms measured, and the precision of the  $^{230}\text{Th}$  analysis at the  $2\sigma$  level (2 standard deviations). This table shows that  $3 \times 10^{10}$  atoms of  $^{230}\text{Th}$  can be measured to  $\pm 2$  ‰ ( $2\sigma$ ) and that  $6 \times 10^8$  atoms of  $^{230}\text{Th}$  can be measured to  $\pm 29$  ‰ ( $2\sigma$ ). The measurements have been verified with replicate analyses on standards and samples (Table 2). The accuracy of U and Th abundances are ultimately based on standard solutions with concentrations which are accurately known from gravimetry (9, 14).

Details about samples can be found in (14). Samples TAN-E-1g, CWS-F-1, and CH-8 have previously been dated by the  $^{14}\text{C}$  method. Sample TAN-E-1g has also been dated by counting of coral growth bands. These samples were analyzed to determine the precision with which very young samples could be dated and to provide a comparison of the ages determined by mass spectrometry and those determined with the other methods. Sample CH-8 was collected along the shore of Lago Enriquillo, a lake in Hispaniola which was thought to be connected to the ocean at the time CH-8 grew (15). TAN-E-1g and CWS-F-1 are from nearshore marine environments.

Samples AFS-10, AFS-11, and AFS-12, AFM-20, and R-52 are from the Rendezvous Hill Terrace on Barbados (16), and E-T-2 and E-L-3 are from Efate Island, Vanuatu (17). Both islands are tectonically active and have been uplifted during the Pleistocene and Holocene. These samples are all thought to have grown during periods of high sea level during the last interglacial period and are known from previous  $^{230}\text{Th}$  dating ( $\alpha$  counting) to have grown between 110 and 140 ky ago (17, 18). Sample OC-51 is from the Worthing Terrace on Barbados (about 85 ky old) and FT-50 is from the Ventnor Terrace, also from Barbados (about 105 ky old) (16),

**Table 1.** Number of  $^{230}\text{Th}$  atoms per analysis, mass of coral analyzed, and  $2\sigma$  uncertainty in  $^{230}\text{Th}$  abundance.

Run	$10^{10}$ atoms of $^{230}\text{Th}$ (No.)	Coral mass (g)	$2\sigma$ uncertainty* (‰)
TAN-E-1g	0.058	2.8	$\pm 29$
CWS-F-1	0.43	4.6	$\pm 8$
CH-8	3.2	4.1	$\pm 3$
OC-51	A	4.4	$\pm 1$
	B	3.8	$\pm 2$
FT-50	A	4.2	$\pm 2$
	B	2.1	$\pm 5$
	C	3.5	$\pm 2$
AFS-10	4.3	0.43	$\pm 3$
AFS-11	4.4	0.41	$\pm 2$
AFS-12	A	2.4	$\pm 3$
	B	3.8	$\pm 2$
	C	2.7	$\pm 2$
AFM-20	3.0	0.28	$\pm 2$
R-52	1.9	0.17	$\pm 3$
E-L-3	2.3	0.26	$\pm 2$
E-T-2	A	2.2	$\pm 2$
	B	2.4	$\pm 2$
VA-1	6.2	0.38	$\pm 3$

\*Errors are based on the standard deviations of the mean of about 60 isotope ratios measured in the course of a mass spectrometric run.

and are thought to have grown during interstadial periods. The Barbados and Vanuatu samples were analyzed in order to determine the precision with which relatively old corals could be dated, to compare these dates to previous results, and to compare the ages to the timing of the peaks in the 65°N insolation curve. Samples AFS-10, AFS-11 and AFS-12 are all from the same locality and were analyzed as a test of the closed-system assumption. Because of their stratigraphic assignment to the same reef crest and proximity to each other (16), they should have similar ages. If they were altered to different degrees, different apparent ages would be recorded by the  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$  chronometer.

VA-1 is from one of the higher terraces in Barbados and has been dated by U-He methods as 520 ky old (16). This sample was analyzed to determine the isotopic characteristics of a very old sample. All the Barbados samples are of the species *Acropora palmata*, which is known to thrive near the crests of coral reefs and tends to live within about 2 m of the sea surface at low tide.

In addition to the coral samples (Table 1), we have also studied K-133, which is a portion of a giant clam (*Tridacna gigantus*) shell collected live from New Guinea in 1977. This was analyzed in order to determine the  $^{238}\text{U}$  and  $^{232}\text{Th}$  concentrations in a modern mollusk and assess the suitability of this material for  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$  dating. The sample size was 2 g for the uranium analysis and 8 g for the Th analysis.

**Measurements and analytical error.** The ages (Table 2) have been calculated from Eq. 1. The errors in age ( $2\sigma$ ) are based on the propagation of analytical errors. Our results demonstrate that very precise ages can be determined for corals 180 years old to 130 ky old (Fig. 1). Fractional errors in age ( $\Delta T/T$ ) as small as 0.005 (at the  $2\sigma$  level) can be obtained for corals that are about  $10^4$  years old. Fractional errors in age smaller than 0.1 can be obtained for corals with ages between 50 and 500,000 years.

The period of time that is pertinent to our test of the astronomical theory is about 130 ky ago. The error in age of a coral which is 130 ky old is about  $\pm 1$  ky (Table 2 and Fig. 1). Since the difference between a high point and a low point on the 65°N insolation curve is about 10 ky, the error in age is small enough to allow a detailed comparison of the times of high sea level and the times of high summer insolation.

Except for CH-8 and VA-1, the  $\delta^{234}\text{U}(T)$  values range from 149 to 174. This spread overlaps the range for open ocean water [140 to 150 (11)] but is somewhat higher. This may reflect the uranium isotopic composition of nearshore waters or possibly a small degree of alteration. Considering errors, however, the  $\delta^{234}\text{U}(T)$  values are consistent with closed-system evolution from seawater and do not show clear evidence for any substantial diagenetic alteration. VA-1 has a  $^{230}\text{Th}/^{238}\text{U}$  ratio higher than the maximum possible value for closed-system evolution from seawater, confirming previous observations (16), and does not permit an age to be calculated. If the U-He age [520 ky (16)] is used to calculate  $\delta^{234}\text{U}(T)$ , a value of  $530 \pm 26$  is determined. This is much higher than the seawater value and is clear evidence for diagenetic addition of  $^{234}\text{U}$ . For CH-8, the value of  $\delta^{234}\text{U}(T)$  is 84, which is much lower than the seawater value. Although the possibility that this sample has been altered cannot be ruled out, it appears that this coral grew in a body of water that was restricted from the open ocean and that the low  $\delta^{234}\text{U}(T)$  value reflects the isotopic composition of this body of water (14, 15).

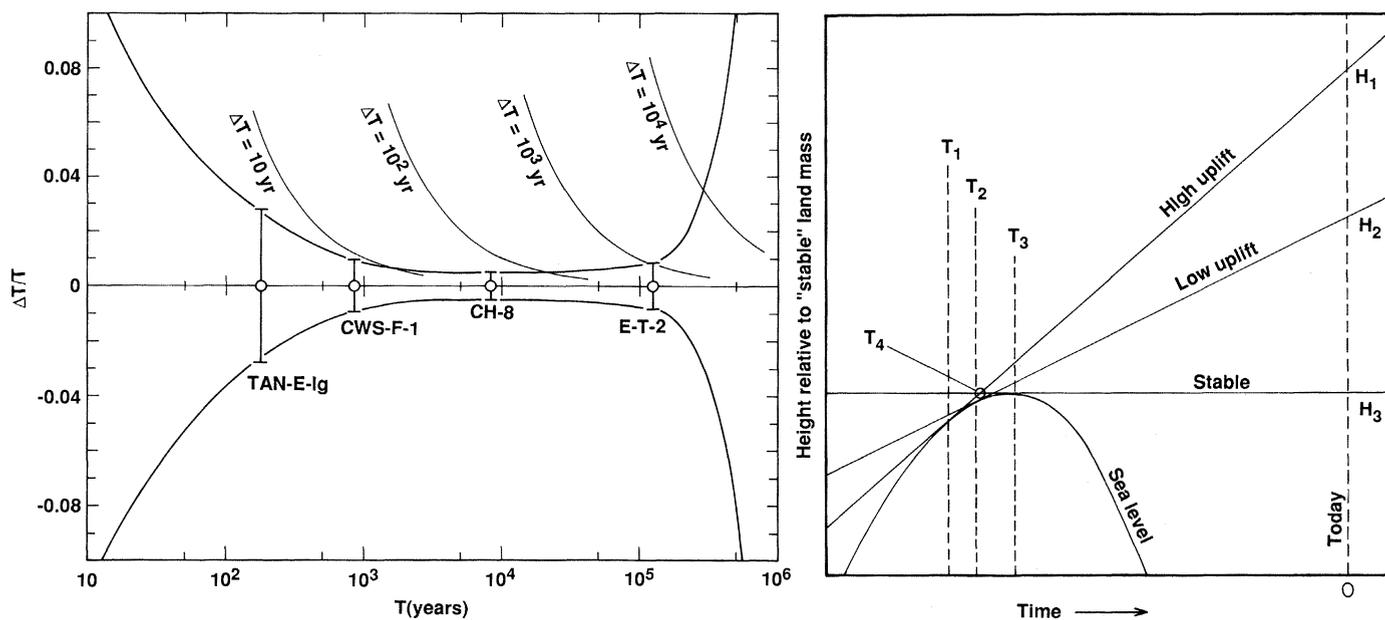
The  $^{232}\text{Th}$  values for corals (Table 1) range from 0.08 to 1.92 pmol/g. Because of the extremely low concentrations and the long mean life of  $^{232}\text{Th}$ ,  $\alpha$ -counting studies have often not reported  $^{232}\text{Th}$ . Examination of published  $^{232}\text{Th}$  concentrations (19) shows that the typical detection limit is about  $10^2$  pmol/g, and values as high as  $10^4$  pmol/g have been reported. The earlier values are at least two orders of magnitude higher than those in Table 1. Our  $^{232}\text{Th}/$

**Table 2.** U and Th isotopic composition and ages of corals. Reported errors are 2 standard deviations. Errors are based on 2 standard deviations of the mean of 60 to 300 isotope ratios measured during a mass spectrometric run.

Sample*	Locality*	$^{238}\text{U}$ (nmol/g)	$^{232}\text{Th}^\dagger$ (pmol/g)	$(^{232}\text{Th}/^{238}\text{U})$ ( $10^5$ ) $^\ddagger$	$\delta^{234}\text{U}(0)^\S$	$\delta^{234}\text{U}(T)^{**}$	$[^{230}\text{Th}/^{238}\text{U}]_{\text{act}}^{\#\#}$	Age $^\dagger\dagger$
TAN-E-1g	Vanuatu	10.80 $\pm$ 0.04	0.083 $\pm$ 0.003	0.77 $\pm$ 0.03	149 $\pm$ 6	149 $\pm$ 6	0.00190 $\pm$ 5	180 $\pm$ 5 yr
CWS-F-1	Vanuatu	10.21 $\pm$ 0.01	0.147 $\pm$ 0.002	1.44 $\pm$ 0.02	152 $\pm$ 4	153 $\pm$ 4	0.00893 $\pm$ 8	845 $\pm$ 8 yr
CH-8	Hispaniola	9.68 $\pm$ 0.01	0.279 $\pm$ 0.002	2.88 $\pm$ 0.02	82 $\pm$ 4	84 $\pm$ 4	0.07951 $\pm$ 28	8294 $\pm$ 44 yr
OC-51	A Barbados	17.05 $\pm$ 0.03	0.17 $\pm$ 0.02	1.0 $\pm$ 0.1	126 $\pm$ 5	161 $\pm$ 6	0.6307 $\pm$ 12	87.5 $\pm$ 0.6 ky
	B	17.36 $\pm$ 0.03	0.15 $\pm$ 0.03	0.9 $\pm$ 0.2	126 $\pm$ 5	162 $\pm$ 6	0.6323 $\pm$ 17	87.9 $\pm$ 0.7 ky
FT-50	A Barbados	13.93 $\pm$ 0.02	0.92 $\pm$ 0.02	6.6 $\pm$ 0.1	126 $\pm$ 5	173 $\pm$ 7	0.7357 $\pm$ 21	112.0 $\pm$ 1.0 ky
	B	13.95 $\pm$ 0.02	0.92 $\pm$ 0.05	6.6 $\pm$ 0.4	127 $\pm$ 4	174 $\pm$ 6	0.7359 $\pm$ 37	111.8 $\pm$ 1.3 ky
	C	13.97 $\pm$ 0.03	0.42 $\pm$ 0.03	3.0 $\pm$ 0.2	124 $\pm$ 5	170 $\pm$ 7	0.7353 $\pm$ 23	112.3 $\pm$ 1.1 ky
AFS-10	Barbados	12.82 $\pm$ 0.02	0.28 $\pm$ 0.02	2.2 $\pm$ 0.2	110 $\pm$ 4	157 $\pm$ 6	0.7728 $\pm$ 24	125.7 $\pm$ 1.2 ky
AFS-11	Barbados	13.67 $\pm$ 0.02	0.74 $\pm$ 0.02	5.4 $\pm$ 0.1	114 $\pm$ 6	161 $\pm$ 8	0.7653 $\pm$ 20	122.6 $\pm$ 1.5 ky
AFS-12	A Barbados	12.66 $\pm$ 0.02	1.40 $\pm$ 0.05	11.1 $\pm$ 0.4	109 $\pm$ 4	153 $\pm$ 6	0.7593 $\pm$ 28	122.1 $\pm$ 1.1 ky
	B	12.62 $\pm$ 0.03	1.57 $\pm$ 0.03	12.4 $\pm$ 0.2	111 $\pm$ 5	157 $\pm$ 7	0.7631 $\pm$ 25	122.7 $\pm$ 1.3 ky
	C	12.43 $\pm$ 0.03	1.37 $\pm$ 0.03	11.1 $\pm$ 0.2	109 $\pm$ 5	155 $\pm$ 6	0.7680 $\pm$ 28	124.5 $\pm$ 1.3 ky
AFM-20	Barbados	13.46 $\pm$ 0.04	1.92 $\pm$ 0.04	14.3 $\pm$ 0.3	111 $\pm$ 4	160 $\pm$ 6	0.7852 $\pm$ 26	129.2 $\pm$ 1.4 ky
R-52	Barbados	13.98 $\pm$ 0.02	0.08 $\pm$ 0.06	0.6 $\pm$ 0.4	115 $\pm$ 6	165 $\pm$ 8	0.7846 $\pm$ 31	128.1 $\pm$ 1.7 ky
E-T-2	A Vanuatu	9.75 $\pm$ 0.01	0.12 $\pm$ 0.03	1.2 $\pm$ 0.3	103 $\pm$ 4	149 $\pm$ 6	0.7811 $\pm$ 17	129.9 $\pm$ 1.1 ky
	B	9.72 $\pm$ 0.01	0.10 $\pm$ 0.03	1.0 $\pm$ 0.3	106 $\pm$ 4	153 $\pm$ 6	0.7811 $\pm$ 12	129.2 $\pm$ 1.1 ky
E-L-3	Vanuatu	11.39 $\pm$ 0.03	0.12 $\pm$ 0.03	1.1 $\pm$ 0.3	113 $\pm$ 5	162 $\pm$ 7	0.7748 $\pm$ 24	125.5 $\pm$ 1.3 ky
VA-1	Barbados	13.37 $\pm$ 0.02	1.08 $\pm$ 0.02	8.1 $\pm$ 0.2	121 $\pm$ 6		1.1940 $\pm$ 39	
K-133	Huon Peninsula, New Guinea (mollusk)	(3.08 $\pm$ 0.08) $\times 10^{-4}$	0.249 $\pm$ 0.002	(8.08 $\pm$ 0.22) $\times 10^4$				
		0.01383 $\pm$ 3	0.00051 $\pm$ 3	3.7 $\pm$ 0.2	140 $\pm$ 5 to 150 $\pm$ 5		$1 \times 10^{-5}$ to $4 \times 10^{-5}$	

\*Further details on samples can be found in (14-18). A, B, and C indicate replicate analyses. For FT-50, A and B are different aliquots of the same dissolved rock fragment; C is a different rock fragment. For AFS-12, A and B are different fractions of the same powder and C is a different rock fragment. For E-T-2 and OC-51, A and B are different rock fragments. All samples are corals except for K-133 which is from a modern giant clam shell.  $^\dagger$ The  $^{232}\text{Th}$  abundance has been corrected for the analytical blank of  $0.02 \pm 0.01$  pmol.

$^\ddagger$ The numbers in this column represent the measured  $^{232}\text{Th}/^{238}\text{U}$  ratio multiplied by  $10^5$ .  $^\S$ The measured uranium isotopic composition calculated as described in text.  $^{**}$ The initial uranium isotopic composition calculated from  $\delta^{234}\text{U}(0)$ , the  $^{230}\text{Th}$  age, and Eq. 2.  $^{\#\#}$ Calculated as described in text with decay constants from (33, 36).  $^\dagger\dagger$ Calculated from Eq. 1; values for the decay constants are from references (33-36).  $^\ddagger\ddagger$  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $\delta^{234}\text{U}(0)$  are from (11). The  $^{230}\text{Th}$  abundance is from surface water values in (10).



**Fig. 1 (left).** Fractional uncertainty in age ( $\Delta T/T$ ) plotted as a function of time, where  $T$  is the age and  $\Delta T$  is the  $2\sigma$  error in age based on the propagation of analytical errors through Eq. 1.  $T$  is plotted on a log scale. Between 180 years and 129.9 ky, the error envelope is drawn through the observed values for several corals. The error envelope is extrapolated to younger and older  $T$  (assuming typical analytical errors). The lowest values of  $\Delta T/T$  are at  $\sim 10^4$  years and values of  $\Delta T/T < 0.10$  can be obtained over the range  $T = 15$  to 500,000 years. Parts of the diagram are contoured in  $\Delta T$ . The intersection of the  $\Delta T = 10^3$  year contour with the error envelope is at  $T \sim 1.3 \times 10^5$  years and indicates that analytical error is small enough at  $T \sim 10^5$  for a critical test of the astronomical theory. **Fig. 2 (right).** Height relative to an arbitrary point on a tectonically stable land mass as a

function of time. A rise and fall of sea level is represented by an arbitrary curve with negative curvature. The lines represent the change in height with time for a specific elevation at locations of high uplift, low uplift, and a tectonically stable locality. The specific elevation is the highest elevation at that locality which is covered by seawater and consequently the highest elevation at a given locality where corals could have grown.  $T_1, T_2,$  and  $T_3$  represent coral ages.  $H_1, H_2,$  and  $H_3$  are present heights of coral terraces. If the assumptions outlined in the text are valid, it may be possible to determine the rate of sea level rise as a function of time. Since both age and present height are measurable quantities, we can test whether the true scenario is consistent with the model.

$^{238}\text{U}$  ratios range from  $0.6 \times 10^{-5}$  to  $14.3 \times 10^{-5}$ . These values are similar to the  $^{232}\text{Th}/^{238}\text{U}$  value for open ocean surface water [ $3.7 \pm 0.2 \times 10^{-5}$  (6)], suggesting that these elements do not fractionate substantially from each other during coral growth. Large shifts in this ratio can therefore be used to indicate Th or U addition during diagenesis or growth from water with  $^{232}\text{Th}/^{238}\text{U}$  different from that found in the open oceans.

The  $^{232}\text{Th}$  concentration of K-133 (a fragment of a modern giant clam shell) is 0.249 pmol/g, which is similar to the value for corals. However, the  $^{238}\text{U}$  concentration is  $3.08 \times 10^{-4}$  nmol/g. This is about  $10^2$  times lower than the lowest previously reported values for a mollusk (20) and  $4 \times 10^4$  times lower than the coral uranium content. The extremely large difference between giant clam and coral uranium contents indicates that these organisms have different mechanisms (or microenvironments) for incorporating uranium into their skeletons. Because of the low  $^{238}\text{U}$  content,  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$  dating of such a material would require a large sample. As giant clams have a mass of about 50 kg, large samples can be found. With the use of mass spectrometric techniques, such an analysis would, in principle, be possible. At secular equilibrium, 200 g of sample would contain  $6 \times 10^8$   $^{230}\text{Th}$  atoms. With our techniques, this number of  $^{230}\text{Th}$  atoms could be measured to an accuracy of  $\pm 3$  percent ( $2\sigma$ ) (Table 1). Although dating of such a sample is now technically feasible, it is clear from previous work (20, 21) that uranium in fossil mollusks is dominantly of diagenetic origin and that the process of uranium uptake must be understood before any serious attempt to date fossil mollusks can be made.

The ages calculated from replicate analyses of AFS-12, E-T-2, FT-50, and OC-51 agree within the  $2\sigma$  error of the measurements. For these samples, replicate analyses were done on different fragments of the same hand specimen. The agreement between the replicate

analyses is not only a demonstration of analytical reproducibility, but also shows that different fragments of the same hand specimen could not have been altered to varying degrees. The results are consistent with the closed-system assumption. The  $^{230}\text{Th}$  age of AFS-11 agrees almost exactly with the ages determined from the three analyses of AFS-12. The age of AFS-10 appears to be slightly greater (1 to 2 ky) than that of AFS-11. Since these samples are from the same outcrop, they should have similar ages as is the case.

**Age comparisons.** The analyzed corals have previously been dated by other methods (Table 3). The younger samples have been dated by  $^{14}\text{C}$  and reported in conventional radiocarbon years (15, 22) and as "corrected ages." For TAN-E-1g and CWS-F-1, the corrected ages were determined from the conventional ages with the use of the calibration curve of Stuiver (23). For CH-8, the corrected age has been given as an approximation since the precise value of the ratio of  $^{14}\text{C}$  to C in the atmosphere at this time does not appear to be known. The corrected  $^{14}\text{C}$  ages do not include adjustments for the natural fractionation of carbon isotopes or for the difference between  $^{14}\text{C}/\text{C}$  in the atmosphere and surface waters. For TAN-E-1g, the corrected age is given by three time intervals since the conventional radiocarbon age intersects the calibration curve three times. TAN-E-1g was collected alive and it was determined from the counting of coral growth bands that the portion of the coral which we analyzed grew between 1804 and 1810 (22). The older samples had been dated by  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$  with the use of  $\alpha$ -counting techniques (17, 18).

Our date for TAN-E-1g of  $180 \pm 5$  years (at the time the sample was analyzed, June 1986) is in excellent agreement with the age determined from counting of growth bands (176 to 182 years old). It also agrees with the corrected  $^{14}\text{C}$  age. Since the conventional radiocarbon age intersects the calibration curve at three points, the corrected  $^{14}\text{C}$  age includes three time intervals that span several

centuries. For CWS-F-1, the date determined by mass spectrometry agrees with the corrected  $^{14}\text{C}$  date within errors. In this case, the conventional radiocarbon age intersects the calibration curve over only one range. For CH-8, there is a substantial difference between the mass spectrometric age and the corrected  $^{14}\text{C}$  age. The reality of this discrepancy requires a more serious comparison of both methods, particularly for samples older than 8000 years, where the availability of samples dated by dendrochronology is limited.

For AFS-10, AFS-11, AFS-12, AFM-20, R-52, FT-50, OC-51, E-L-3, and E-T-2, the agreement between the mass spectrometrically determined ages and the earlier ages determined by  $\alpha$  counting generally appears to be good at the  $2\sigma$  level. The error in age, based on analytical errors, for the mass spectrometric measurements is five to ten times smaller than the error in age for the  $\alpha$ -counting measurements.

**Rate of sea level rise between 129 and 122 ky.** The response of a coral reef relative sea level rise was first discussed by Darwin (24). He observed that reef-building corals thrive at water depths of less than 30 m and reasoned that if sea level were to rise, the reef must grow upward to remain near the sea surface or die. When sea level stops rising, the upward growth of the coral reef would stop. As sea level falls, the crest of the coral reef would remain as a subaerially exposed coral terrace. The age of the coral at the highest point of the terrace would represent the time at which sea level reached its highest elevation.

The situation at Barbados is more complicated. The Rendezvous Hill Terrace has present elevations of 27 to 61 m above sea level (16) (Table 4). Terraces of similar age (about 120 ky) in areas which may have been tectonically stable over the last 120 ky have elevations about 6 m above sea level (25). The difference in elevation between the Rendezvous Hill Terrace and the terraces from these areas is inferred to be due to tectonic uplift on Barbados that has occurred over the past 120 ky (2) (Table 4). The Barbados terraces must be

**Table 4.** Present height, age, and average uplift rate of Barbados terraces.

Sample	Present height above sea level* (m)	Age (ky)	Rate of uplift† (m/ky)
AFS-11	30	122.6	0.20
AFS-12	30	123.1	0.19
AFS-10	30	125.7	0.19
R-52	37	128.1	0.24
AFM-20	55	129.2	0.38

\*From topographic maps in reference (16); errors are about  $\pm 3$  m. †Minimum uplift rates calculated as described in text.

the net result of two processes: fluctuations in sea level and tectonic uplift. This concept has been discussed by Mesollela *et al.* (2) for Barbados and by Bloom *et al.* (26) for New Guinea. Further discussion of the relation of reef morphology and zonation to sea level changes can be found in (27, 28).

It is necessary to distinguish between changes in the absolute height of sea level and tectonic displacements. Consider the following simple model. We assume that the sea level curve has negative curvature over a pertinent time range and that the rate of tectonic uplift at a given locality is constant (Fig. 2). The curve labeled "sea level" represents the change in sea level with time. The lines represent tectonic uplift for a specific elevation at localities with different uplift rates. The specific elevation is the one that intersects the sea level curve at the tangent point. This is the highest elevation that is covered by seawater and, consequently, is the highest elevation where a coral reef could have grown. The age of this coral records the time at which sea level was rising at the same rate as the land mass. For a "stable" land mass, this age ( $T_3$ ) represents the time when sea level reached its maximum height. For a rising land mass, this age ( $T_1$  or  $T_2$ ) is older than the sea level maximum. The model

**Table 3.** Coral ages determined by different methods or techniques. Ages refer to the ages in 1986. Reported errors are  $2\sigma$ . For  $^{14}\text{C}$  and  $\alpha$  counting, the errors are based on counting statistics. Errors in mass spectrometric ages are based on the standard deviation of the mean of 60 to 300 isotope ratios measured in the course of a mass spectrometric run.

Sample	$^{14}\text{C}$ * (conventional) (years)	$^{14}\text{C}$ † (corrected) (years)	Ring counting‡ (years)	$^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$ § ( $\alpha$ counting) (ky)	$^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$ (mass spectrometric)
TAN-E-1g	270 $\pm$ 120	30 to 70, 180 to 270, or 300 to 500	176 to 182		180 $\pm$ 5 years
CWS-F-1	980 $\pm$ 120	780 to 1010			845 $\pm$ 8 years
CH-8	8990 $\pm$ 120	~10,000			8294 $\pm$ 44 years
OC-51 A				85 $\pm$ 4	87.5 $\pm$ 0.6 ky
B					87.9 $\pm$ 0.7 ky
FT-50 A				107 $\pm$ 7	112.0 $\pm$ 1.0 ky
B					111.8 $\pm$ 1.3 ky
C					112.3 $\pm$ 1.1 ky
AFS-10				123 $\pm$ 8	125.7 $\pm$ 1.2 ky
AFS-11				127 $\pm$ 9	122.6 $\pm$ 1.5 ky
AFS-12 A				129 $\pm$ 9	122.1 $\pm$ 1.1 ky
B					122.7 $\pm$ 1.3 ky
C					124.5 $\pm$ 1.3 ky
AFM-20 A				117 $\pm$ 8	129.2 $\pm$ 1.4 ky
B				127 $\pm$ 9	
R-52 A				107 $\pm$ 6	128.1 $\pm$ 1.7 ky
B				120 $\pm$ 9	
E-T-2 A				141 $\pm$ 16	129.9 $\pm$ 1.1 ky
B					129.2 $\pm$ 1.1 ky
E-L-3				141 $\pm$ 16	125.5 $\pm$ 1.3 ky

\* $^{14}\text{C}$  ages are as reported in (15, 22) in radiocarbon years; the mean life of 8033 years was used; no corrections have been made for natural fractionation of carbon isotopes, the difference between  $^{14}\text{C}/\text{C}$  in surface water and the atmosphere, or differences in initial  $^{14}\text{C}/\text{C}$ . † $^{14}\text{C}$  ages have been corrected by us to dendroyears using the curves of Stuiver (23) for TAN-E-1g and CWS-F-1 and for CH-8, assuming a  $^{14}\text{C}/\text{C}$  initial ratio from Klein *et al.* (37) for tree rings ~8000 years old. No corrections have been made for natural fractionation of carbon isotopes or the difference between  $^{14}\text{C}/\text{C}$  in the surface water and the atmosphere. ‡From reference (22). §Ages from reference (18) except for E-T-2 and E-L-3 which are from reference (17).

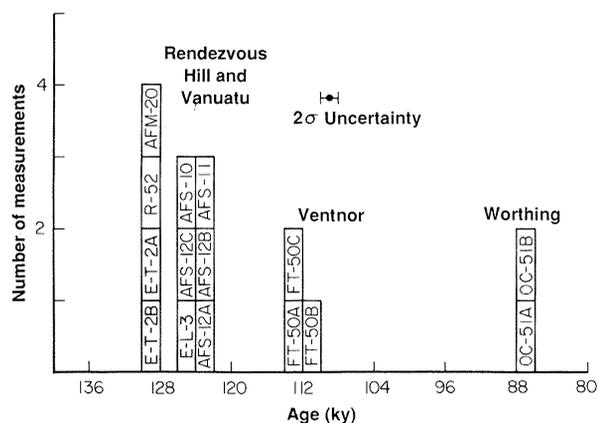
predicts that older corals should have higher present heights ( $T_1 > T_2 > T_3$  for  $H_1 > H_2 > H_3$ ). By dating corals from localities with different uplift rates, one could, in principle, use this model to determine the rate of sea level rise as a function of time.

To use this approach, we must know the uplift rate at each locality. Exact uplift rates cannot be calculated from the present heights ( $H_1, H_2, H_3$ ) and ages ( $T_1, T_2, T_3$ ), but bounds can be placed on this rate. At the "high uplift" locality, the uplift rate is given by  $(H_1 - H_3)/T_4$ .  $T_4$  is not known, but must lie between  $T_1$  and  $T_3$ . Therefore, the uplift rate lies between  $(H_1 - H_3)/T_1$  and  $(H_1 - H_3)/T_3$ .

For the Rendezvous Hill Terrace on Barbados, the corals with older ages have higher present heights as required by the model (Table 4). One of the Vanuatu samples does not meet this requirement. The Vanuatu samples have not been included in this analysis since their stratigraphic relationships are not clearly understood. Also listed are uplift rates calculated by subtracting 6 m ( $H_3$ ) (25) from the present height ( $H_1$  or  $H_2$ ) and dividing by the age ( $T_1$  or  $T_2$ ). These are lower bounds on the uplift rate. Upper bounds could be calculated with the use of  $T_3$ . Mass spectrometric values for  $T_3$  have not been obtained. Values for  $T_3$  from  $\alpha$  counting are, within error, the same as our values for  $T_1$  and  $T_2$  (25). Therefore, the minimum uplift rate is very close to the true uplift rate and is taken to represent this quantity.

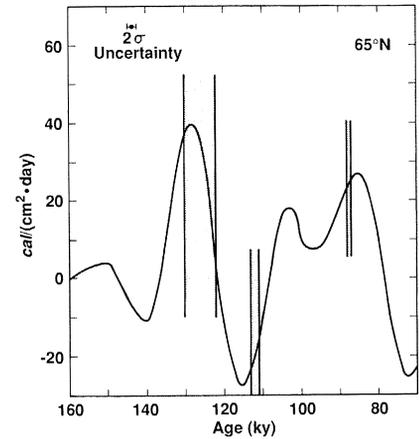
If the assumptions used in deriving the above model are valid for the Barbados samples, then the uplift rate must be equal to the rate of sea level rise at the time of coral growth. It appears that the rate of sea level rise at 129 ky was about 0.4 m/ky, and between 126 and 122 ky the rate was about 0.2 m/ky. These rates of sea level rise suggest that sea level did not rise by more than 2 or 3 m between 129 and 122 ky. The extension of the present database to stable localities and localities with very high uplift rates should provide further insight into this problem.

**The Milankovitch curve and the specific ages of corals.** A histogram of our data for the Barbados and older Vanuatu samples (Fig. 3) shows three distinct age ranges. As the Barbados samples were collected from three distinct terraces, this would be expected. The Vanuatu samples apparently grew at the same time as the Rendezvous Hill corals from Barbados. The ages of these samples range from 130 to 122 ky. Analyses of one sample from the Ventnor Terrace and one sample from the Worthing Terrace on Barbados give ages of 112 ky and 87.7 ky, respectively. On the basis of the



**Fig. 3.** The ages of corals from Barbados and Vanuatu. The typical  $2\sigma$  error in age for each analysis is shown by the error bar. The Barbados samples are from three different terraces. The Vanuatu samples are stippled. These data suggest that the last interglacial period lasted from 130 ky to at least 122 ky ago and that interstadial periods occurred 112 and 87.7 ky ago. We have found no evidence for high sea level just prior to 130 ky ago.

**Fig. 4.** The vertical axis represents the average solar insolation received at the top of the atmosphere at 65°N for the summer half year minus the present value. The curve is drawn for  $T = 160$  ky to 70 ky (29). The stippled bands represent the ages shown in the histogram in Fig. 3. The oldest band represents the range of ages for a number of corals from the Rendezvous Hill Terrace in Barbados and from Vanuatu. The other bands represent the age and  $2\sigma$  error in age for two individual samples from the Worthing and Ventnor terraces in Barbados. The error bars represent the typical  $2\sigma$  errors in age for the mass spectrometric measurements. The oldest range of ages coincides with an insolation high and lends support to the Milankovitch hypothesis.



model for reef growth, the three age ranges should represent times when sea level was close to a maximum value but was still rising (at the rate of tectonic uplift).

The three age ranges (Fig. 3) have been superimposed on a graph (Fig. 4) of the summer solar insolation received at 65°N latitude as a function of time [calculations by Berger (29)]. This curve has low values between 166 and 140 ky, rises to a very high value at 128 ky and drops to a very low value at 116 ky. It has maxima at 103 ky and 85 ky, then drops to a low value at 73 ky. The oldest range of coral ages occurs almost exactly at the time of the insolation maximum at 128 ky. The oldest corals in this range may have grown as much as 2 ky before the insolation high; the youngest in this range grew as much as 6 ky after the insolation high. Previous data, both geomorphic and radiometric ( $\alpha$  counting) (30), have suggested the possibility that sea level was high about 140 ky ago. This would predate the 128 ky insolation peak by more than 10 ky and could not be the direct result of orbital forcing. We have found no evidence for high sea level 140 ky ago. Our oldest range of ages supports the idea that Pleistocene sea level highs can be the result of orbital forcing. The apparent high position of sea level 130 ky ago requires that, as the insolation curve increases, the phase lag between the insolation and sea level curves is small. The high position of sea level 6 ky after the insolation peak suggests that as the insolation curve decreases, the phase lag is significantly larger. From the range of ages, it appears that sea level was high for at least 8 ky during the last interglacial period.

The youngest coral (87.7 ky) grew about 2 ky before the insolation high at 85 ky. The apparent high position of sea level at this time could be the result of orbital forcing. However, the 112-ky-old coral grew 9 ky before the 103 ky insolation high. The high position of sea level at this time does not appear to coincide with an insolation high.

We conclude that the use of mass spectrometric techniques for the measurement of  $^{230}\text{Th}$  and  $^{234}\text{U}$  in corals permits a substantial increase in the range over which useful ages can be determined with the  $^{238}\text{U}$ - $^{234}\text{U}$ - $^{230}\text{Th}$  system with a range from about 50 years to about 500 ky. The precision with which ages can be determined, based on analytical errors, is about five to ten times higher than by  $\alpha$ -counting methods. The sample size requirement is about 30 times smaller. The precise determination of  $\delta^{234}\text{U}$  and  $^{232}\text{Th}$  will also permit a clearer evaluation of possible open-system behavior. The analyses on young corals suggest that this approach may be prefera-

ble to  $^{14}\text{C}$  analysis for these materials. The basic problems of diagenesis and element remobilization will remain the fundamental problems that require study. Our precise measurements on corals that grew during the last interglacial period lie specifically within the 128-ky Milankovitch peak and support the idea that major changes in Pleistocene climate can be caused by orbital forcing. The age of a coral that grew during an interstadial period also supports this view. However, the age of a coral that grew during another interstadial period suggests that some changes in Pleistocene climate may be the result of other mechanisms. The ability to analyze small samples is very important with regard to studies of marine terraces. Reef-forming corals are largely limited to the tropics; however, solitary corals grow in marine environments worldwide, but only occur in small masses. Few terraces have yielded enough solitary coral for  $\alpha$ -counting analysis. When the techniques outlined here are used, the small sample size will also allow dating of well-preserved solitary corals. If foraminifera could be dated, the absolute chronology for the  $^{18}\text{O}/^{16}\text{O}$  record in deep sea sediments could be determined. The uranium concentration in foraminifera has been reported as 0.11 nmol/g (31). At secular equilibrium, 1 g of foraminifera contains  $1.1 \times 10^9$  atoms of  $^{230}\text{Th}$ , which could be measured to an accuracy of  $\pm 1$  percent ( $2\sigma$ ) by means of mass spectrometric techniques. These techniques have also been used in our laboratory to measure the concentrations of  $^{230}\text{Th}$  ( $10^7$  atoms per liter) and  $^{232}\text{Th}$  in 1 to 3 liters of seawater (11, 32).

#### REFERENCES AND NOTES

- W. S. Broecker *et al.*, *Science* **159**, 297 (1968).
- K. J. Mesollella, R. K. Matthews, W. S. Broecker, D. L. Thurber, *J. Geol.* **77**, 250 (1969).
- H. H. Veeh and J. M. Chappell, *Science* **167**, 862 (1970).
- M. M. Milankovitch, *Canon of Insolation and the Ice Age Problem* (Königlich Serbische Akademie, Beograd, 1941). English translation by the Israel Program for Scientific Translations, Jerusalem, 1969.
- W. T. Hyde and W. R. Peltier, *J. Atmos. Sci.* **42**, 2170 (1985).
- W. R. Peltier, in *Irreversible Phenomena and Dynamical Systems Analysis in Geosciences*, C. Nicolis and G. Nicolis, Eds. (Reidel, Dordrecht, 1987), p. 399.
- J. D. Hays, J. Imbrie, N. J. Shackleton, *Science* **194**, 1121 (1976).
- J. Imbrie *et al.*, in *Milankovitch and Climate*, A. L. Berger, J. Imbrie, J. Hays, G. Kukla, B. Saltzman, Eds. (Reidel, Dordrecht, 1984), p. 269.
- J. H. Chen and G. J. Wasserburg, *Anal. Chem.* **53**, 2060 (1981).
- Y. Nozaki, Y. Horibe, H. Tsubota, *Earth Planet. Sci. Lett.* **54**, 203 (1981).
- J. H. Chen, R. L. Edwards, G. J. Wasserburg, *ibid.* **80**, 241 (1986).
- A. Kaufman and W. S. Broecker, *J. Geophys. Res.* **70**, 4039 (1965).
- R. H. Steiger and E. Jäger, *Earth Planet. Sci. Lett.* **36**, 359 (1977).
- R. L. Edwards, J. H. Chen, G. J. Wasserburg, *ibid.* **81**, 175 (1987).
- F. W. Taylor, P. Mann, S. Vaestro, Jr., K. Burke, *J. Geol.* **93**, 311 (1985).
- M. L. Bender *et al.*, *Geol. Soc. Am. Bull. Part I* **90**, 577 (1979).
- A. L. Bloom, C. Jouannic, F. W. Taylor, appendix to R. P. Ash, J. N. Carney, A. Macfarlane, *Geology of Eface and Offshore Islands, New Hebrides Geol. Surv. Regional Report* (1978).
- T.-L. Ku, in preparation.
- J. F. Marshall and B. F. Thom, *Nature (London)* **263**, 120 (1976).
- W. S. Broecker, *J. Geophys. Res.* **68**, 2817 (1963).
- A. Kaufman, W. S. Broecker, T.-L. Ku, D. L. Thurber, *Geochim. Cosmochim. Acta* **35**, 1155 (1971).
- F. W. Taylor, written communication.
- M. Stuiver, *Radiocarbon* **24**, 1 (1982).
- C. R. Darwin, *The Structure and Distribution of Coral Reefs* (Smith and Elder, London, ed. 2, 1874).
- T.-L. Ku, M. A. Kimmel, W. H. Easton, T. J. O'Neil, *Science* **183**, 959 (1974).
- A. L. Bloom, W. S. Broecker, J. M. A. Chappell, R. K. Mathews, K. J. Mesollella, *Quat. Res.* **4**, 185 (1974).
- R. K. Mathews, *ibid.* **2**, 368 (1972).
- P. Aharon and J. Chappell, *Paleogeogr. Paleoclimatol. Paleocol.* **56**, 337 (1986).
- A. L. Berger, *Quat. Res.* **9**, 139 (1978).
- W. S. Moore, in *Uranium Series Disequilibrium: Applications to Environmental Problems*, M. Ivanovich and R. S. Harmon, Eds. (Oxford Univ. Press, New York, 1982), p. 481.
- T.-L. Ku, *J. Geophys. Res.* **70**, 3457 (1965).
- A. J. Spivack, J. H. Chen, G. J. Wasserburg, *Trans. Am. Geophys. Union* **67**, 1066 (1986).
- J. W. Meadows, R. J. Armani, E. L. Callis, A. M. Essling, *Phys. Rev. C* **22**, 750 (1980).
- M. Lounsbury and R. W. Durham, in *Proceedings International Conference on Chemical Nuclear Data, Measurements and Applications*, M. L. Hurrell, Ed. (Institute of Civil Engineers, London, 1971), p. 215.
- P. de Bievre *et al.*, *ibid.*, p. 21.
- A. H. Jaffey, K. F. Flynn, L. E. Glendenin, W. C. Bentley, A. M. Essling, *Phys. Rev. C* **4**, 1889 (1971).
- J. Klein, J. C. Lerman, P. E. Damon, E. K. Ralph, *Radiocarbon* **24**, 103 (1982).
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