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Uranium-Series Ages of Pacific Atoll Coral

Abstract. The thorium-230: uranium-234 method of dating corals and oolites has been evaluated in detail for reliability, and various criteria have been established. Reliable ages for extensive coral formations of about 6000 and 120,000 years were obtained. A hiatus in the development of coral between 6000 and 120,000 years ago on the Pacific atoll of Eniwetok implies that conditions did not permit coral growth during this period. The record prior to 120,000 years ago is not clear, probably because of a lack of unaltered samples.

Barnes et al. (1) first demonstrated the feasibility of using Th²³⁰, the daughter of U²³⁸, for dating marine carbonates; they showed that living corals contain several parts per million (ppm) of U^{238} almost completely free of its product, Th²³⁰. Fossil coral from progressively greater depths in borings from Eniwetok yielded progressively higher Th²³⁰:U²³⁸ ratios, approximating the equilibrium ratio at depths beyond 30 m. This discovery suggested the possibility that Pleistocene marine carbonates in the range of a few thousand to a few hundred thousand years old might be dated by this technique.

Subsequent studies (2), while substantiating the initial work, emphasized that recrystallization was evidence that the chemical system had not remained closed. Both this and other work (3)suggested that the presence of Th²³² was evidence of possible incorporation of external Th^{230} . Thurber's report (4) that the dissolved uranium in sea water is enriched in U²³⁴ activity by 15 percent over that of its parent U²³⁸ not only required a revision of previous age estimates but also opened the possibility of an independent age method based on the decay of the excess U^{234} .

Comparisons between Th²³⁰ and C¹⁴ ages, although limited to samples less than 25,000 years old, provided an obvious means of establishing the validity of both methods. The relation between the concentration of Ra²²⁶ and that of its parent Th²³⁰ offered a means of checking the assumption that the system had remained closed for the past few thousand years. Our report is of a cooperative effort between Washington University and Columbia University groups to apply these approaches in evaluating Th²³⁰ ages obtained on carefully selected coral samples from borings made in the mid-Pacific atolls (5). Fanale and Schaeffer (6) report analyses of He:U in several of these samples as an independent means of establishing their absolute age.

The quantitative relations between the four isotopes U^{238} , U^{234} , Th^{230} , and Ra²²⁶ as a function of age are given in Figs. 1 and 2 (7). The initial ratios of U²³⁴:U²³⁸, Th²³⁰:U²³⁴, and Ra²²⁶:U²³⁴ are taken respectively as 1.15, .00, and .09 (see below). The sys-

tem is assumed to have been closed to further addition or removal of these isotopes; the equations used to generate these graphs are given by Broecker (8).

The measurements reported here were made as follows. Th²³⁰ and Th²³² were measured by alpha-counting thin sources of separated thorium. The Washington University group used alpha-decay curve analysis and alpha spectrometry (2, 9); the Lamont group, alpha spectrometry (10). Uranium measurements were made by either fluorometry (8, 11) or alpha spectrometry. Ra²²⁶ measurements were made by alpha counting the Ra²²² daughter (8); C^{14} measurement, by proportional counting of CO₂ gas (12). The percentage of calcite was determined by x-ray analysis.

In Table 1 the ratios of one nuclide to another are relative decay rates rather than mass ratios. Two nuclides at radioactive equilibrium will have an activity ratio of unity. The first and last samples in Table 1 are control samples: one living and one very ancient coral. Ratios observed for the living coral are consistent with previous estimates (2, 3, 13); they are used to establish the initial conditions listed above. As expected, the Miocene coral shows equilibrium throughout the uranium decay series. Based on the results on living coral and on the assumption that the system has remained closed, the following criteria can be used to evaluate the results on samples of unknown age.

1) The sample should contain less than a few percent calcite. As coral forms as pure aragonite, the presence of calcite indicates that recrystallization or cementation has taken place; such processes are likely to lead to violation of the closed-system assumption.

2) The uranium concentration in the



Fig. 1. Theoretical decay and growth curves for U^{234} and Th^{230} in a fossil marine coral.

sample should be about 2 to 3 ppm. Empirical information suggests that corals do not distinguish between Ca, Sr, Ra, and U during growth (2, 8, 14), and coral ordinarily has a U:Ca ratio similar to that of sea water. There is no evidence that this situation changed during Pleistocene times; thus fossil coral with uranium concentrations outside this range may well have suffered serious chemical alteration; recrystallized coral, for example, often is low in uranium (2, 11).

3) The $U^{234}:U^{238}$ ratio, corrected for age, should be $1.15 \pm .03$. Uranium in sea water contains 15 percent more U^{234} than U^{238} activity; no significant variations have appeared with geographic location or depth (15, 16). If this excess has prevailed during the last several hundred thousand years, the ratio observed in any fossil coral (corrected for decay by use of the Th²³⁰: U^{234} age) should be 1.15. If the assumptions are true, any sample yielding a divergent ratio is suspect.

4) The Ra^{226} : Th²³⁰ ratio should be consistent with the age of the sample. Samples with ages greater than 70,000



Fig. 2. Predicted decay and growth curves for Ra^{226} : Th^{230} in a fossil marine coral.

years should yield ratios of unity within the experimental error (Fig. 2). Those between 7000 and 70,000 years of age should reflect the lag in the growth of Ra^{226} , and those less than 7000 years of age should reflect not only this lag but also the influence of the residual initial Ra^{226} . If corals form with negligible Th^{230} and with 10 percent of the equilibrium value of Ra^{226} , departure of the measured ratio from that predicted in Fig. 2 indicates that the system has been open within the last 7000 years. Unfortunately, chemical alteration that occurred more than 7000 years ago will leave no imprint on this ratio.

5) The Th²³⁰:Th²³² ratio in the sample should exceed 20. Because of the very low concentration of Th²³² in sea water, measurable quantities of this isotope would not be expected in coral, and such is the case (Table 1). Thus the presence of Th²³² in fossil corals may be taken as evidence of secondary addition. Since the Th²³⁰:Th²³² ratio in natural waters commonly ranges from 1 to 4 (15), each unit of Th^{232} added would be accompanied by at least one unit of Th²³⁰. Because the uncertainty in the ratio of these two isotopes makes a Th²³² correction undesirable, in our work any coral sample with a Th²³⁰:Th²³² ratio less than 20 is considered suspect.

6) The Th²³⁰:U²³⁴ age for the sample should be consistent with its C¹⁴ age. The samples reported here fall in two quite different age categories: those 3000 to 6000 years old and those greater than 25,000 years in age. For

Table 1. Complete data requ ired to calculate and evaluate reliability of ages of Pacific coral samples.

L No.	USGS No.	Depth (m)	Analyzed by	Calcite (%)	U ²³⁸ (ppm)	Th ²³² :U ²³⁸ †	U ²³⁴ :U ²³⁸ †	Th ²³⁰ :U ²³⁴ †	Ra ²²⁶ :U ²³⁴ †
584A	Loc. 15	Surface	L	<1	$3.3 \pm .3^{*}$		$1.15 \pm .01$		$0.09 \pm .01$
			L		$3.25 \pm .09$	$0.003 \pm .001$	$1.12 \pm .01$	$0.004 \pm .001$.10 ±.01
482A	MU-7-4	3.9 to 7.2	W		$2.5 \pm .1^{*}$			$.042 \pm .008$	
			L	2	$2.5 \pm .3^{*}$				$.035 \pm .003$
482B	MU-7-10	7.2 to 7.8	W		$3.2 \pm .1^*$			$.061 \pm .003$	
			L	3	$2.8 \pm .3^{*}$		$1.15 \pm .01$		$.053 \pm .004$
725A	MU-7-11	7.2 to 7.8	L	<2	$2.72 \pm .03$	$.002 \pm .001$	$1.14 \pm .01$	$.046 \pm .003$	$.037 \pm .003$
482C	MU-7-12	10.2 to 10.8	W		$3.1 \pm .1^*$			$.053 \pm .005$	
			L	2	$3.1 \pm .3^*$.041 ± . 003
482D	MU-7-13	14.4 to 15.3	W		$2.9 \pm .1^{*}$	$.001 \pm .001$		$.58 \pm .05$	
			L	<1	$2.98 \pm .03$		$1.08 \pm .01$	$.66 \pm .02$	$.63 \pm .06$
482E	MU-7-23	19.2 to 20.7	W	-	$2.7 \pm .1^*$			$.67 \pm .05$	
			L	2	$2.6 \pm .3^{*}$		$1.09 \pm .01$.62 ±.06
725B	MU-7-23	19.2 to 20.7	L	<2	$2.74 \pm .03$	$.002\pm.001$	$1.11 \pm .02$	$.68 \pm .06$	$.63 \pm .06$
482F	MU-7-34	24.0 to 25.5	W		$2.7 \pm .1^*$			$.79 \pm .06$	
			L	<2	$2.32 \pm .03$	$.003 \pm .001$	$1.09 \pm .01$	$1.02 \pm .04$	$1.00 \pm .05$
			L		$2.40 \pm .05$	$.004 \pm .001$	$1.05 \pm .02$	$1.02 \pm .04$	$0.92 \pm .07$
802A	MU-7-32	24.0 to 25.5	L	20	$2.95 \pm .06$	$.020 \pm .005$	$1.05 \pm .02$	$0.92 \pm .06$	$.84 \pm .08$
482G	MU-7-41	27.0 to 29.1	W		$2.5 \pm .1^*$			$.79 \pm .06$	
			L	1	$2.4 \pm .3^{*}$		$1.08 \pm .01$.90 ±.06
725C	MU-7-41	27.0 to 29.1	L	10	$1.96 \pm .02$	$.003 \pm .001$	$1.06 \pm .01$	$.82 \pm .04$.80 ± .08
614B	RU-2-14	14.1 to 15.6	W		$2.5 \pm .1^{*}$			$.69 \pm .05$	
			L	_	$3.0 \pm .3^{*}$		$1.09 \pm .01$.61 ±.06
			L	7	$2.79 \pm .04$	$.002 \pm .001$	$1.11 \pm .02$	$.73 \pm .04$	$.66 \pm .05$
725E	3	15.9 to 17.4	L	<1	$2.64 \pm .02$	$.002 \pm .001$	$1.09 \pm .01$	$.64 \pm .04$	$.62 \pm .06$
802B	EN-6-31	30.0	L	<1	$2.32 \pm .04$	$.020 \pm .005$	$1.03 \pm .02$.91 ±.04	$.95 \pm .07$
725D	EN-9-45	33.0	L	25	$2.22 \pm .02$	$.004 \pm .001$	$1.06 \pm .01$	$.98 \pm .04$.91 ± .07
802D	AO-1-23	42.0	L	5	$4.45 \pm .04$	$.005 \pm .001$	$1.00 \pm .01$	$.91 \pm .04$	$.98 \pm .05$
			Ľ	_	$4.36 \pm .04$	$.001 \pm .001$	$1.01 \pm .01$	$.93 \pm .05$	$1.00 \pm .05$
725F	2-9-13	47.4 to 48.9	L	8	$3.63 \pm .04$	$.043 \pm .005$	$1.06 \pm .01$	$.75 \pm .03$	0.94 ±. 07
	~~		L		$3.37 \pm .04$	$.041 \pm .005$	$1.06 \pm .03$	$.84 \pm .03$	$.93 \pm .05$
802C	K-1-16	51.0	L	4	$3.31 \pm .03$	$.002 \pm .001$	$1.13 \pm .01$.87 ±.05	.77 ±. 06
614A	F-1-1	51.0 to 57.3	W		$3.1 \pm .1^*$			$.93 \pm .07$	
			L		$2.7 \pm .3^{*}$		$1.07 \pm .01$.98 ±. 08
			L	30	$3.20 \pm .06$	<.001	$1.08 \pm .01$.98 ± .03	.98 ± . 06
584B	F-1	210.0	L	26			$1.00 \pm .01$		
			L	4	$2.11 \pm .03$	$.007 \pm .001$	$1.01 \pm .01$	$1.00 \pm .06$	$1.00 \pm .06$

L, Lamont Geological Observatory; W, Washington University; USGS, U.S. Geological Survey. * U analyses by fluorometry. \ddagger Isotope ratios are in rates of relative decay.

Table 2. Characteristics of coral samples from less than 12 meters below the surface, Eniwetok.

USGS No.	Depth (m)	$rac{{ m Th}^{230*}}{{ m Th}^{232}}$	$\frac{Ra^{226} \text{ obs.}}{Ra^{226} \text{ pred.}}$	Th ²³⁰ age (yr)	C ¹⁴ age (yr)	
Loc. 15	Surface	0.5		400 ± 100	<200	
MU-7-4	3.9 to 7.2		0.8	4500 ± 900	3800 ± 150	
MU-7-10	7.2 to 7.8		1.2	6600 ± 400	5600 ± 150	
MU-7-11	7.2 to 7.8	23	1.0	5000 ± 300	5000 ± 150	
MU-7-12	10.2 to 10.8		1.0	5700 ± 600	5900 ± 150	
Obs, observed;	pred. predicted; USG	S, U.S. Geo	logical Survey.	* Activity ratio.	······································	

Table 3. Characteristics of samples from 12 to 21 meters below the surface, Eniwetok.

USGS No.	Depth (m)	Th ²³⁰ : Th ²³² *	${ m Ra}^{226}$: Th $^{230}*$	Th ²³⁰ age (10 ³ yr)	(U ²³⁴ :U ²³⁸) ₀ *	C ¹⁴ age (10 ³ yr)	He ⁴ age [corr. (6)] (10 ³ yr)
MU-7-13 MU-7-23 MU-7-23 RU-2-14 3	48-51 64-69 64-69 47-52 53-58	660 340 365 320	$\begin{array}{c} 1.00 \pm .11 \\ .92 \pm .13 \\ .92 \pm .13 \\ .90 \pm .13 \\ .97 \pm .13 \end{array}$	$ \begin{array}{r} 100 \pm 10 \\ 115 \pm 15 \\ 120 \pm 15 \\ 130 \pm 15 \\ 110 \pm 10 \end{array} $	$\begin{array}{c} 1.11 \pm .01 \\ 1.12 \pm .01 \\ 1.15 \pm .03 \\ 1.14 \pm .02 \\ 1.12 \pm .02 \end{array}$	33	120

USGS, U.S. Geological Survey; corr., corrected. * Activity ratio.

those in the first category there is little reason to believe that the C^{14} ages could deviate from the absolute by more than 20 percent. However, because recent contamination amounting to a few percent by weight can give a very ancient carbonate an apparent age in the 25,000- to 40,000-year range, finite C^{14} ages in the second category can be considered as little more than lower limits.

7) The Th^{230} : U^{234} ages should be consistent with stratigraphic data. Because many of the samples reported here are in vertical sequence, such comparisons are useful.

Agreement in the radiochemical analyses between the two laboratories is excellent except in one instance. The Th²³⁰:U²³⁴ ratio determined at Washington University for sample 482-F is significantly lower than the Th²³⁰:U²³⁴ and Ra²²⁶:U²³⁴ ratios determined at Lamont. It should be pointed out that only the two analyses made at Lamont are true duplicates, since the interlabo-

ratory split entailed slicing one large specimen rather than grinding and quartering. As the Th^{230} concentrations measured are identical, the difference in the $Th^{230}:U^{234}$ ratio may stem from the difference in the uranium results.

The reported analytical uncertainties are derived principally from counting statistics. Replicate determinations of standard samples suggest that these truly represent the standard deviation and that systematic errors are absent.

Table 2 contains ages computed for coral samples with ages less than 6000 years. Except for the living coral they are cuttings from drill hole MU-7, Mujinkarikku Island, Eniwetok Atoll (5). The results fit the above criteria very well. The C¹⁴ and Th²³⁰:U²³⁴ ages are in surprisingly good agreement. Only sample 482-B appears to have more Th²³⁰ and Ra²²⁶ than its C¹⁴ age and stratigraphic position would indicate.

Table 3 contains ages computed for

Table 4.	Characteristics	of samples	from below	24 meters,	Eniwetok.
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Calcite (%)	Analyzed by	USGS No.	Depth (m)	Th ²³⁰ :Th ^{232*}	Ra ²²⁶ :Th ^{230*}	Th ²³⁰ age (10 ³ yr)	U ²³⁴ age (10 ³ yr)	He ⁴ age [corr. (6)] (10 ³ yr)
20	L	MU-7-32	80-85	46	$.91 \pm .10$	>220	400 ± 150	175
<1	L	MU-7-34	80-85	300	$.94 \pm .05$	>220	275 ± 50	
	W					160 ± 30		
1	L	MU-7-41	90-97		$1.14 \pm .10$	210†	225 ± 35	
	W					160 ± 30		
10	L	MU-7-41	90-97	200	$.98 \pm .09$	175 ± 25	325 ± 75	
<1	L	En-6-31	100	450	$1.04 \pm .08$	225 ± 40	550 ± 200	340
25	L	En-9-45	110	250	$.93 \pm .08$	>220	325 ± 75	
5	L	AO-1-23	140	180	$1.07 \pm .05$	235 ± 40	>700	400
8	L	2-9-13	158-168	20	$1.18 \pm .06$	165 ± 10	325 ± 50	
4	L	K-1-16	170	435	$.89 \pm .08$	200 ± 40	50 ± 25	900
30	L	F-1-1	170-191	>1000	$1.00 \pm .07$	>250	240 ± 20	400
	W					>200		

USGS, U.S. Geological Survey; corr., corrected. * Activity ratio. † Based on Ra²²³ rather than Th²³⁰ measurement. 2 JULY 1965

samples from between 12 and 21 m, which all apparently formed close to 115,000 years ago; analytical data for these samples are in very good agreement. The average U^{234} : U^{238} ratio. corrected for age, is somewhat lower than that in present day sea water; whether this low ratio reflects temporal variations in U²³⁴:U²³⁸ in sea water or is related to analytical uncertainties in measurements of the U^{234} : U^{238} ratio is unknown. The radiocarbon age of 33,000 years obtained for sample 482-E illustrates the danger in assigning finite radiocarbon ages to old carbonates: even 2 percent of modern carbon can give this sample its apparent radiocarbon age.

The samples reported on in Table 4 cannot be readily assigned ages; although from different cores, and therefore possibly not in stratigraphic sequence, many of them do not even show internal consistency.

The Th²³⁰-U²³⁴ ages have so much scatter that calculated initial U²³⁴:U²³⁸ ratios would probably have little meaning. Therefore U²³⁴:U²³⁸ ages have been calculated on the assumption of an initial ratio of 1.15, in order to compare these with the U-He ages.

The Th²³⁰-U²³⁴ ages in this range are very sensitive to small changes in the relative concentrations of uranium and thorium. The difficulty in assigning ages from the Th²³⁰ data may stem from the fact that the analytical precision available does not enable precise calculation of ages when the Th²³⁰ is near equilibrium. At the same time, the calcite found in most of these samples may indicate that the system has been opened enough to disturb the normal relations between the nuclides of the uranium series. In some instances —K-1-16 and AO-1-23, for example—

the inconsistencies are most likely due to chemical alteration.

The Th²³⁰ method of dating coral appears to be valid for samples obeying the seven criteria specified. It would appear that assignment of ages in this set of samples is limited to samples less than 150,000 years, either by the analytical precision now available or by the fact that truly unaltered samples have not been used.

Because of the considerable scatter in the age-corrected U^{234} : U^{238} values. the validity of ages based on the disappearance of the excess U²³⁴ remains to be demonstrated. The He:U dating method, discussed by Fanale and Schaeffer (6), should be very valuable in future evaluations of both the Th²³⁰: U^{234} and the $U^{234}: U^{238}$ methods.

The presence of coral-bearing horizons aged 120,000 years in the lagoons of mid-Pacific atolls strongly suggests that sea level and climate at that time resembled modern conditions. The absence of horizons aged between 5000 and 100,000 years suggests (2) that the ocean level was considerably lower during this period than at present, causing a hiatus in coral deposition. The temptation to use this information to establish the ages of one or more of the Pleistocene interglacials must be subdued pending more absolute age information.

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Uranium-Series Dating of Corals and Oolites from Bahaman and Florida Key Limestones

Abstract. Samples of fossil coral and oolite from marine formations of the Bahamas and the Florida Keys have been analyzed for their concentrations of uranium and thorium isotopes. Ages calculated from the ratio Th^{230} : U^{234} clearly indicate periods of marine limestone formation at about 85,000, 130,000, and 190,000 years ago, implying that the surface of the ocean was then close to its present level.

The uranium-series method of dating, as applied to Pleistocene coral from mid-Pacific atolls, has been described in some detail (1); we refer the reader to that paper for discussion of the method and the establishment of criteria used in judging the reliability of these ages. These criteria may be summarized as follows: (i) the samples should show no evidence of recrystallization; (ii) the uranium content should ordinarily be about 2.7 parts per million (ppm); (iii) the activity ratio of Th²³⁰ to Th²³² should exceed 20; (iv) the activity ratio of Ra²²⁶ to Th²³⁰ should be $1.0 \pm .1$; (v) the activity ratio of U234 to U238, corrected for decay of U^{234} , should be 1.15 ± .02; (vi) the ages should agree with independent age estimates; and (vii) the ages should be consistent with stratigraphic information.

Table 1 contains data on recent corals in the Florida Keys. The U^{234} : U²³⁸ ratio is about that found in sea water; scatter of the data on the U^{234} : U²³⁸ ratio probably results from analytical uncertainty. The concentrations of Th²³² and Th²³⁰ are low, as one would expect from their low concentrations in sea water (2). Similar uranium concentrations and ratios of Ra²²⁶: U²³⁸, Th^{232} : U²³⁸, Th^{230} : U²³⁸, and U²³⁴: U²³⁸ have been reported for currently forming oolite (3, 4).

Table 2 summarizes results of tests on samples of fossil oolite and coral from the Florida Keys and the Bahamas; this work complements that of Osmond et al. (4).

Newell (5) finds Bahaman deposits representing at least two distinct sealevel stands near that of present-day sea level in formations above a wellconsolidated basement of marine limestone. Two formations of eolianite dunes of marine oolite are separated by a weathered zone. Terraces, veneered with beach deposits containing coral and molluses, are cut into the lower formation.

The samples from the Berry Islands, Bahamas, for which ages have been determined, include two corals. One sample, from the basement rock, shows an age of 200,000 years. The other sample, the younger, is from one of the terraces, and gives an age of 80,000 years. Both samples satisfy the criteria, and the ages are supported by He-U ages determined by Fanale and Schaeffer (6).

Two samples of the dune rock have been analyzed, one from the younger eolianite (717M) and one from the older (717N); both contain a large fraction of calcite. However, the other criteria are satisfied for the younger dune, and the age of 140,000 years may be judged reliable. The other sample may be judged unreliable on the basis of its low Ra226 : Th230 ratio and high initial U²³⁴ : U²³⁸ ratio. This sample, while forming a reasonable stratigraphic sequence with the other samples, cannot be assigned ages based on the Th²³⁰: U²³⁴ ratio at present with any certainty.

The oolite sample from Bimini appears to have a reliable age of 155,000